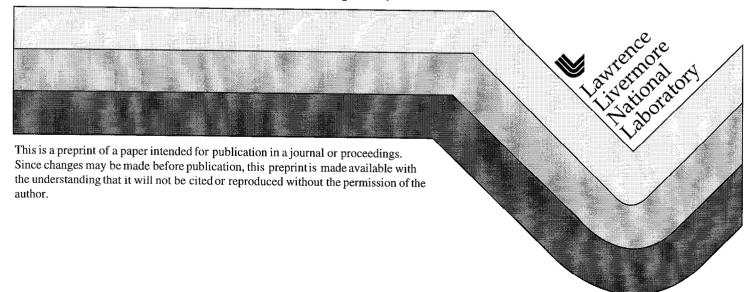
From Separations to Reconstitution— A Short History of Plutonium in the US and Russia

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FROM SEPARATIONS TO RECONSTITUTION — A SHORT HISTORY OF PLUTONIUM IN THE US AND RUSSIA

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ABSTRACT

During the cold war plutonium was produced in reactors in both the US and Russia. It was then separated from the residual uranium and fission products by a variety of precipitation processes, such as Bismuth Phosphate, Redox, Butex, Purex, etc. in the US and uranium acetate and Purex in Russia. After a period of time in the field, plutonium weapons were recycled and the plutonium re-purified and returned to weapons. Purification was accomplished by a variety of aqueous and molten salt processes, such as nitric-hydrofluoric acid dissolution followed by anion exchange, Purex modifications, molten salt extraction, electrorefining, etc. in the US and nitric acid dissolution or sodium hydroxide fusion followed by anion exchange in Russia. At the end of the Cold War, plutonium production of weapons-grade plutonium was cut off in the US and is expected to be cut off in Russia shortly after the turn of the century. Now both countries are looking at methods to reconstitute plutonium with fission products to render it no longer useful for nuclear weapons. These methods include immobilization in a ceramic matrix and then encasement in fission product laden glass, irradiation of MOX fuel, and disposal as waste in WIPP in the US and irradiation of MOX fuel in Russia. This paper will detail the contrast between the treatment of plutonium during the cold war and after the cold war was over.

-1- 4/15/99

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INTRODUCTION

Glenn T. Seaborg^[1,2] stated that "plutonium is unique among the chemical elements:

- It is a synthetic element, the first realization of the alchemist's dream of large-scale transmutation;
- It was the first synthetic element produced in visible amounts;
- It has unusual and interesting chemical and metallurgical properties;
- It is one of the most dangerous poisons that man must learn to handle;
- It was discovered and methods for its production were developed in secrecy during World War II;"
- Its discovery was inextricably intertwined with the discovery and study of nuclear fission;
- Its naming was inextricably intertwined with the naming of the outer planets; and
- Production of weapons-grade plutonium has been inextricably intertwined with the politics of the "Cold War.

Since its discovery and its dramatic emergence at Nagasaki, plutonium has altered the course of history, changed the concepts and consequences of war, and paradoxically has become a powerful instrument for peace. With the ending of the "Cold War", production of weaponsgrade plutonium has stopped in the United States. Production has been curtailed in Russia^[3]; three dual-purpose reactors still produce weapons-grade plutonium — two at Tomsk-7 (renamed Seversk) and one at Krasnoyarsk-26 (renamed Zheleznogorsk Mining and Chemical Combine). In a 1994 United States-Russian agreement that has yet to enter into force, Russia agreed to close the remaining operating reactors by the year 2000.

DISCOVERY OF PLUTONIUM[1,2,4]

Beginning in the mid-1930's, spurred by the discovery of the neutron by Chadwick in 1932 and artificial radioactivity by the Joliot-Curies in 1934, the new breed of nuclear scientist (including both chemists and physicists) became intrigued with the prospect of synthesizing new "artificial" elements not found in nature. Between 1934 and 1939, several dozen research papers described the discovery and study of transuranium elements with Z=93, 94, 95, and 96. For example, Fermi and coworkers began experiments in 1934 to try to produce element 93 by bombarding uranium with neutrons to give ²³⁹U which they expected would beta decay to produce ²³⁹93. Based on the then accepted periodic table, they believed element 93 should behave like the lighter homologs manganese and rhenium. They performed a chemical separation designed to separate element 93 with a precipitate of rhenium sulfide and found a 13-minute activity which in a paper published in 1934^[5] they tentatively assigned to element 93. Ida Noddack (a discoverer of rhenium) published a paper on after in which she expressed her doubt about their assignment and suggested that the bombarded nuclei might have disintegrated into several larger fragments! In subsequent years, Hahn, Meitner, and Strassmann^[7] in Berlin seemed to confirm these results and reported other isotopes of eka-rhenium as well as of eka-osmium. iridium, and -platinum. However, there were many unsettling aspects about these results and in 1938 Curie and Savitch^[8] found a 3.5-hour activity with the chemical properties of a rare earth,

-2- 4/15/99

but they were unable to explain this and it was at first greeted with some skepticism by the Berlin group. Finally, Hahn and Strassmann decided to perform their own experiments and found several new activities that they attributed to radium and actinium isotopes and isomers. Meitner (now in Stockholm) at least, found these result unsatisfying there were now some 16 new species that originated with ²³⁸U, including multiple isomerism, and beta-decay energies which were too high for the measured half-lives! Furthermore, the yields were markedly enhanced when slow neutrons were used, making the postulated (n,α) reactions to make thorium which then β -decayed to Ra (Ac) isotopes energetically impossible. Meitner urged that they re-examine the experiments and it was then that Hahn and Strassmann^[9] began the series of experiments which culminated in the discovery of nuclear fission by showing that the activities attributed to Ra, Ac, and Th actually followed the chemistry of Ba, La, and Ce! Thus, all of these "transuranium elements" were, if fact, products of nuclear fission with atomic numbers below 60. But they still hesitated to believe their results, and stated, "As nuclear chemists, being in some respects close to physics, we have not yet been able to take this leap, which contradicts all previous experiences in nuclear physics. It could still perhaps be that a series of unusual coincidences have given us deceptive results."

However, during the Christmas holidays of 1938, Meitner together with her nephew, physicist Otto Robert Frisch, who had come to visit her in Sweden, unraveled the puzzle. They worked out all the essential features of nuclear fission based on the liquid drop model and the masses involved. Upon returning to Copenhagen, Frisch spoke with Niels Bohr, Danish physicist, who immediately agreed with their interpretation and they proceeded to submit a letter to Nature on "Disintegration of Uranium by Neutrons: A New Type of Nuclear Reaction" [10]. In this elegant paper, in just over a page, they outlined the essentials of the theory of nuclear fission, including the total energy released and the recoil energy of the fission fragments, which Frisch [11] quickly detected experimentally.

NEPTUNIUM (Z=93) AND PLUTONIUM (Z=94)

Now there were no more transuranium elements, as Fermi's transuranium elements could all be attributed to fission products which had obscured the activity of the new element 93. This led to new experiments in 1940. Seemingly, all of the nuclear scientists in the world then set out to investigate the new phenomenon. In Berkeley, Edwin M. McMillan, while investigating the recoil of fission products produced in the irradiation of a thin layer of ²³⁸U with slow neutrons, found a 2.3-day activity which did not recoil out and appeared to be the daughter of 23-minute ²³⁹U. In the spring of 1940 he and Philip H. Abelson chemically separated and identified ^[12] the new element Np.

$$^{238}U + n \longrightarrow ^{239}U + \gamma$$

$$^{239}U \xrightarrow{\beta, 23 \text{ min utes}} ^{239}Np \xrightarrow{\beta, 2.3 \text{ days}} ^{239}Np$$

It was obvious to them that the new element should beta decay to the same mass number of element 94, but they were unable to identify it. Bombardments were begun with deuterons in the 60-Inch Cyclotron at Berkeley, but McMillan was called away and Glenn T. Seaborg wrote to

-3- 4/15/99

ask him whether he and his graduate student Arthur C. Wahl and a fellow instructor Joseph W. Kennedy should carry on this research. He agreed and in December 1940 they detected alpha decay of an activity which grew in to the chemically separated neptunium fraction and had a half-life estimated to be about 50 years.

$$^{238}U + ^{2}H \longrightarrow ^{238}Np + 2n$$

$$^{238}Np \xrightarrow{\beta} ^{239}Pu (\sim 50 \ years)$$

They sent a short note to Washington describing their results. But they didn't consider they had sufficient proof to claim discovery until finally on the night of February 23, 1941, Art Wahl oxidized it with peroxydisulphate and Ag ion and it was shown to have properties similar to uranium but not osmium as suggested earlier. This was also communicated to Washington, but these two articles were not published until 1946^[13,14]. Soon after, a 1.2 kg sample of uranyl nitrate was irradiated with neutrons in the 60-Inch cyclotron for two days in order to produce enough ²³⁸Np so that after its purification it would decay to form 0.5 µg of ²³⁹Pu whose half-fife was calculated to be about 30,000 years. Shortly after in March, 1941, it was shown that its fission cross section with slow neutrons was about 50% greater than for ²³⁵U, which was later published by Kennedy, Seaborg, Segré, and Wahl^[15]. This information formed the basis for the US wartime Plutonium Project at the Metallurgical Laboratory of the University of Chicago whose mission was to develop a method for the production of Pu in quantity and for its chemical separation on a large scale. Elements 93 and 94 were name neptunium (Np) and plutonium (Pu), because uranium was originally named after Uranus, and Neptune and Pluto are the next planets beyond Uranus, hence, the names neptunium and plutonium. (For more details on these discoveries and naming, see G.T. Seaborg, [16,17])

PLUTONIUM PRODUCTION IN THE UNITED STATES

During World War II and the ensuring Cold War, the US developed a massive industrial complex to research, produce, and test nuclear weapons^[18]. During Manhattan Project, there was a crew of more than 100,000; 37 installations in 13 states; and more than a dozen university laboratories^[18]. At the height of the "Cold War", the "nuclear weapons complex" included nuclear reactors, chemical processing plants, laboratories, fabrication and maintenance facilities that manufactured tens of thousands of nuclear warheads, and conducted more than 1000 nuclear explosive tests. At its peak, the nuclear weapons complex consisted of 16 major facilities including vast reservations of land in the states of Idaho, Nevada, South Carolina, and Washington. Its diversity ranged from isolated tracts of desert in Nevada and Islands in the South Pacific where weapons were tested, to warehouses in downtown New York City that once stored uranium.

Between 1944 and 1988, the United States built^[19] and operated 14 plutonium-production reactors, nine at Hanford and five at Savannah River, producing approximately 100 metric tons of plutonium. Plutonium separations plants, at Hanford and Savannah River, supported these reactors. Weapons parts were fabricated in Colorado, Florida, Missouri, Ohio, and Tennessee. Final assembly was in Texas. National Laboratories in California and New Mexico supported the Nuclear Weapons Complex.

-4- 4/15/99

Most of the uranium for the Manhattan Project^[19] came from rich deposits in Africa and Canada, but more that 400 mines eventually opened in the United states, primarily in Arizona, Colorado, New Mexico, Utah, and Wyoming. (For each kilogram of plutonium produces, miners must take approximately 1,000 tons of uranium ore from the ground.)

It is reported^[19] that Nobel Prize-winning, Danish physicist Niels Bohr had argued that building an atomic bomb "can never be done unless you turn the United States into one huge factory." Years later, he told his colleague Edward Teller, "I told you it couldn't be done without turning the whole country into a factory. You have done just that."

Weapons production stopped^[19] in the United States the late 1980's, initially to correct widespread environmental and safety problems, and was later ended indefinitely because of the end of the Cold War. This stoppage left a legacy of thousands of contaminated areas and buildings, and large volumes of "backlog" waste and special nuclear material residues requiring treatment, stabilization, and disposal. Approximately one-half million cubic meters of radioactive high-level, mixed, and low-level waste must be stabilized, safeguarded, and dispositioned, including a quantity of plutonium sufficient to fabricate thousands of nuclear weapons.

PLUTONIUM PRODUCTION IN RUSSIA

Work toward an atomic bomb^[20] began in the USSR in 1939 when Igor Kurchatov alerted the Soviet government to the possible military significance of nuclear fission. Kurchatov suspected that fission research might be under way in Nazi Germany. Soviet physicists realized in 1940 that the United States must have also been pursuing a big secret program when:

- 1. There was no American response to the June 1940 *Physical Review* report on the observed rare spontaneous fissioning in uranium.
- 2. The names of prominent physicists, chemists, metallurgists and mathematicians disappeared from international journals; secrecy itself gave the secret away.

The German invasion of the USSR in June 1941 temporarily ended the Russian program. Research toward a uranium weapon seemed too far removed from the immediate necessities of war.

But the Soviet government assembled an advisory committee. The committee endorsed atomic bomb research and recommended Kurchatov to head it. Early in 1943 work was resumed in Moscow. Nuclear scientists were recalled from the war front, from industry, for the research institutes that had been evacuated to the rear. They did experiments and made theoretical calculations concerning the reactions involved in both nuclear weapons and nuclear reactors, they began work designed to lead to the production of suitable pure uranium and graphite, and they studied various possible means for the separation of uranium isotopes.

-5- 4/15/99

THE EARLY YEARS

Historically, precipitation processes^[21] were first used in the recovery of plutonium. Although the plutonium concentration may be too small to exceed the solubility product of even very insoluble compounds, plutonium can be removed from solution by co-precipitation. In general, plutonium will co-precipitate if the anion of the bulk precipitate forms an insoluble plutonium compound with plutonium in a particular oxidation state.

In the early years the US used the bismuth phosphate process [21,22,23] which is based on the ability of a BiPO₄ precipitate to carry Pu (IV) but not Pu (III) or Pu (VI). The USSR used sodium uranyl acetate, [24,25,26,27] a specific carrier for the MO_2^{2+} ion. Thus, Pu (VI), but not Pu (III) or Pu (IV) is carried when $NaUO_2(C_2H_3O_2)_3$ is precipitated from solution. The US also used the precipitation of $NaPuO_2(C_2H_3O_2)_3$ in the final purification stages before the conversion of plutonium to metal.

United States

When Brigadier General Leslie R. Groves^[28, 29, 30] assumed command of the Manhattan Engineer District on September 17, 1942 he was reportedly horrified at the major uncertainties which faced the plutonium project. No one had yet built a pile that had gone critical and been controlled. There were various potential methods for cooling the reactor in which the uranium would be irradiated. The means of subsequent chemical separations was unknown. Only the week^[17] before had 2.77 µg of plutonium been isolated. The equipment needed to do the job had not been designed. The scientist could not guess to "within a factor of ten" the amount of plutonium that was needed to make each atomic bomb. Nevertheless, it was Groves' philosophy that "nothing would be more fatal to success than to try to arrive at a perfect plan before taking any important step." He could tackle the practical questions: What materials would be needed? Which contractor had any experience that could help? Where would the plant be located?

During November 1943, E. I. du Pont de Nemours & Company was signed as the prime contractor^[28,29,30] to construct and operate the plutonium production plant. For purely legal reasons, provisions were made in the contract for du Pont to receive a fee of one dollar. A dusty tract of land lying between the towns of White Bluffs, Hanford, and Richland, Washington was chosen as the site in January 1943 and the Secretary of War issued a formal directive on February 8, 1943, authorizing acquisition of approximately one-half million acres of land. B-Reactor,^[30,31] the world's first large-scale plutonium production reactor, became operational in September 1944. D-Reactor became operational in December 1944 and F-Reactor in February 1945.

Since the plutonium was to be used for weapons, the only chemical processing required was the extraction of plutonium, free from fission-products contamination and from the spent natural-uranium fuel. In 1943, several methods were proposed; the method selected was a precipitation process. [17,28] There were a number of reasons for this choice. Since the preliminary chemical studies were made with microgram quantities of plutonium, the choice was based primarily open the belief that a precipitation method would be the one most likely to be successful when transferred from the laboratory to the production plant scale. The precipitation process involved operations (precipitation and dissolution) that were very

-6-

familiar to chemical engineers. These few operations, which were applied repeatedly, and which required only simple equipment made possible a simplification in the design, manufacturing, and operating problems.

The separation plants were designed when only sub-microscopic quantities of plutonium existed. The two flowsheets that seemed to offer the best prospects employed virtually the same equipment and piping layout, so that it was possible to go ahead with a design. Most of the major design decisions for Hanford had to be made before the Clinton pilot plant reactor was in operation. Each building [32] was 800 feet long, 65 feet wide and 80 feet tall, pouredconcrete structures so massive the workers called them Queen Marys; the British ocean liner of that name was only a fifth again as long. The containment building were essentially concrete boxes in which there were individual cells containing the various pieces of equipment involved in the process. To provide protection from the intense radioactivity, the cells were surrounded by concrete walls seven feet thick and were covered by six feet of concrete. Each building contained 40 cells. A standard equipment group occupied two cells: a centrifuge, a catch tank, a precipitator and a solution tank, all made of stainless steel. The solutions moved through these units by steam-jet syphoning, a low-maintenance substitute for pumps. There were three necessary steps to the separation process: dissolution, precipitation, and centrifugal removal of the precipitate. These were repeated from equipment group to equipment group down the canyon of the separations building. The end products were radioactive wastes, stored on site in underground tanks, and small quantities of highly purified plutonium nitrate. T-Plant, the world's first large-scale plutonium separation facility, processed its first batch of irradiated fuel rods from B-Reactor on December 26, 1944. Apparently, the fuel was cooled only about 40 days^[33] to allow the ²³⁹Np to decay away (half-life of 2.3 days) so that the ²³⁹Np would not add additional radiation to the plutonium product. Batch sizes were limited to no more than 375 grams of plutonium.

The chemical process, called the bismuth-phosphate process, was a batch operation^[1,17,33] that used repeated dissolution, precipitation, metatheses, digestion, agitation and centrifugation to achieve a high degree of separation of plutonium from uranium and associated fission products. First the aluminum-silicon "cans" or jackets that surrounded the fuel rods was dissolved by slowly adding 50% NaOH to a hot solution of 26% NaNO₃. The fuel slugs themselves were dissolved using 60% HNO₃. After adjustment of the solution to about 50% uranyl nitrate hydrate (UNH), the uranyl ion was masked with sulfate and plutonium was converted to tetravalent state with NaNO₂. Then the plutonium separated by means of on BiPO₄ carrier precipitation — first Bi(NO₃)₃ was added and then H₃PO₄ was added over a three-hour period. The precipitate, containing plutonium and fission products was centrifuged and washed with water and then dissolved in HNO₃. The plutonium oxidized to Pu (VI) using NaBiO₃ and Na₂Cr₂O₇ and prepared for precipitation with bismuth, ceric, and zirconium scavenger. After collection by centrifugation and washing with water, the cake was again dissolved in HNO₃. Fission products were then separated by treatment with ammonium fluosilicate, reduced with ferrous ammonium sulfate, and then precipitated by the addition of bismuth nitrate and phosphoric acid. After collection of the product by centrifugation and water washing, the precipitate was dissolved in nitric acid. Additional decontamination from uranium was achieved by adjusting to the tetravalent state, treating via scavenger precipitation, the ammonium fluosilicate treatment, and reprecipitating the plutonium by BiPO₄ carrier precipitation. The cake was again dissolved in nitric acid. After the gross gamma activity associated with the product was reduced to a level of 100 mr/hour

-7- 4/15/99

at a distance of ten feet from a 300-gallon batch, the product was transferred from the canyon to the finishing line. Then the solution was treated to remove the bulk of the bismuth and some additional activity. After additional decontamination cycles, with the plutonium alternating between the tetravalent and hexavalent state, plutonium was then coprecipitated with LaF₃. This product was metathesized with KOH and dissolved in nitric acid. Separation of plutonium and lanthanum was accomplished by two plutonium peroxide precipitations. Seaborg^[32] notes proudly, "The yields in the first plant runs ... ranged between 60 and 70 per cent and reached 90 per cent early in February 1945."

The final precipitate was dissolved in nitric acid and shipped to Los Alamos where it underwent the final processing steps that transformed it into plutonium metal. Most of the shipments —small subcritical batches in metal containers in wooden boxes — traveled in convoy by Army ambulance via Boise, Salt Lake City, Grand Junction and Pueblo to Los Alamos. [32]

The batch process is inherently inefficient and has the additional disadvantage of losing the uranium to the waste stream. Nonetheless, the wisdom of selecting the bismuth phosphate initially was substantiated by later events, such as the fact that development of the plant-scale Redox process required the efforts of 100 scientist and engineers for more than three years.

Russia

During the creation of the Soviet nuclear industry in the 1940s, scientist at the Khlopin Radium Institute in Leningrad (now St. Petersburg) developed the technology of plutonium extraction from irradiated fuel. ^[24,31] The first fuel reprocessing method was based on slightly soluble sodium uranyl acetate precipitation from nitric acid solutions.

The uranium^[23,24,25,26,27] fuel or targets were dissolved in nitric acid, yielding a solution that was about 1 to 2 molar nitric acid. An oxidant such as sodium dichromate was added to the mother liquor to adjust the plutonium valence to Pu (VI). The solution was then adjusted to near a neutral solution of about 5 M NaNO₃-1 M CH₃COOH- 0.5 M Na (CH₃COO), and the uranium and plutonium allowed to co-precipitate. The precipitate is allowed to settle in conical tanks and the bulk of the solution, containing the fission products, was then decanted to another tank. The precipitate was apparently washed with a similar solution and decanted as given above. During the first years of plant operation (1949 to mid-1950s), acetate-nitrate solutions made up the bulk of radioactive high level waste. [25,26,27] Apparently, these waste had sodium nitrate concentrations exceeding 100 g/L and sodium acetate concentrations of 60 to 80 g/L. These solutions occupied a large volume, were difficult to store, and due to their high salinity, concentration by evaporation was impossible.

The slurry was then apparently dissolved and the precipitation repeated as many times as necessary to reduce the radiation level.

The plutonium [24, 25, 26, 27] was then separated from the uranium by first reducing the plutonium to either Pu (III) or Pu (IV) and again adjusting the solution as above to precipitate only the "slightly" sodium uranyl acetate salt. The plutonium solution was apparently decanted and the uranium slurry again dissolved and precipitated as above to decontaminate the uranium.

The plutonium^[24] in the solutions was oxidized to Pu (VI) and the sodium plutonyl acetate precipitated. Sodium plutonyl acetate is a pink compound. [23]

-8- 4/15/99

Just as with the bismuth phosphate process, the precipitation is very slow. [25, 26, 27]

From a variety of sources it appears as if the Russians then dissolved the plutonium in nitric acid; then neutralized the solution with ammonium carbonate and added BaCl₂ to precipitate a mixture of double salts: Ba₃(PuO₂)₂(CO₃)₅(H2O)₁₀, BaPuO₂(CO₃)₂(H₂O)₃, Ba₃(PuO₂)₂(CO₃)₃ (OH)(H₂O)₆, etc. Taking this up into molten BaCl₂ would presumably give PuCl₃ that would then be reduced by calcium metal to plutonium metal.

PLUTONIUM PRODUCTION AFTER WORLD WAR II

Shortly after World War II, relations between the United States and the Soviet Union really began to deteriorate. On February 9, 1946, Joseph Stalin gave a speech at the Bolshoi Theater that marks the beginning of the deterioration in relations with the US. On March 5, 1946, Winston Churchill delivers "iron curtain" speech at Fulton, Missouri. Then in March 1946, President Truman "declared" the "Cold War." The most enduring legacy of the "Cold War" was the arms race.

In the United States, the nuclear arms race resulted in the development of a vast research, production, and testing network that come to be known as the "nuclear weapons complex." Russia also built its network of "closed cities."

United States

At Hanford, the reduction oxidation (REDOX) processing plant, [22, 28, 31] the first continuous solvent extraction plant in the world, was completed and began operations in January 1952. REDOX used methly isobutyl ketone (known as hexone) as the organic extractant, and aluminum nitrate as the "salting agent." Hanford used tall "packed columns" to achieve contact between the organic and aqueous phases of the process.

The Purex process uses a mixture of tributyl phosphate (TBP) and a hydrocarbon diluent to extract uranyl nitrate and tetravalent plutonium nitrate from an aqueous solution containing nitric acid. The Purex process was suggested by the discovery of J. C. Warf^[22, 34] in 1949 that tetravalent cerium could be separated from the nitrates of trivalent rare earth's by solvent extraction with TBP. The Purex process was developed by the Knolls Atomic Power Laboratory^[34, 35] of the General Electric Company and carried through the pilot-plant stage at Oak Ridge National Laboratory from 1950 to 1952. E. I. du Pont de Nemours and Company adopted it for the Savannah River plutonium-production plant. The Purex process was put into operation in F-Canyon in November 1954. Its success there led to replacement of the Redox process by the Purex process at Hanford in January 1956.

The Commissariat á l'Energie Atomique^[22] developed a solvent extraction process similar to Purex using TBP for use in the French plutonium separation plant at Marcoule. Since then, the Purex process has replaced the Butex process at Windscale, has been used in Argentina, Belgium, Brazil, Germany, Italy, India, Japan, and the USSR; it has become the universal choice for separation of uranium and plutonium from fission products in irradiated uranium.

-9-

4/15/99

Russia

In the Soviet Union there was no plutonium to process until after the war. They built three facilities^[3, 20, 24, 25, 26, 27, 31] for the processing of plutonium — two of the plants were in Siberia and the third in the Southern Ural Mountains:

- 1. The Siberian Chemical Combine (five dual-purpose reactors, two reactors are still operating) at Tomsk-7. The Tomsk-7 complex is located on the Tom River; it is about 15-km north-northwest of the Siberian City of Tomsk in Tomsk Oblast.
- 2. The Mining and Chemical Combine (Three dual-purpose reactors, one reactor is still operating) at Krasnoyarsk-26. This site is located near the Stolba preserve, 64 km from Krasnoyarsk, and has had several names such as "Devyatka," Krasnoyarsk-26, Zheleznogorsk, and Atomgrad. The site was built the 1950s next to the Yenisey River. A road leads to a tunnel at the base of a mountain, where the nuclear station is located underground at a "depth of 250 meters. More than 65,000 prisoners and 100,000 soldiers were required to dig the underground areas.
- 3. The Mayak Chemical Combine (five reactors) at Chelyabinsk-65. The Chelyabinsk-65 complex, located about 70 km north of the city of Chelyabinsk in Russia is the site of the first production reactor complex built in Russia. The Production Association Mayak has carried out nuclear activities since 1948 at the Chelyabinsk-40 site. The site referred to as Chelyabinsk-40 is located near the towns of Kyshtym and Kasli and is about 70 km north of the city of Chelyabinsk. Chelyabinsk-65 was used to refer to a town of 83,000 built to house the Mayak staff, is located 12 km from the site. The name Chelyabinsk-40 was dropped about 1991.

The site is located on generally flat terrain among numerous lakes, marshes, and floodplains of several rivers. Intermediate level waste was discharged directly to the Techa River from the reprocessing plant from 1949 to 1951.

These reprocessing plants continued to use the acetate precipitation process for some time. [24,25,26,27] To process the acetate-nitrate radioactive solutions, a precipitation-crystallization-sorption technology was developed by Spitzin of the Physical Chemistry Institute of the USSR Academy of Sciences. [25, 26, 27] This process solved three problems:

- 1. Radionuclides were concentrated (by a factor of 100 based on volume) by precipitation of insoluble compounds having a large sorption capacity for fission products
- 2. Recovery of acetate-ion for recycling
- 3. Production of high purity crystalline sodium nitrate, which could be used as a fertilizer or for producing caustic soda.

Radionuclide concentration was achieved by coprecipitation with low-solubility compounds such as iron and chromium hydroxides, iron and nickel sulfides and nickel ferrocyanide. Ruthenium and strontium were "concentrated" on nickel and chromium hydroxides; zirconium, niobium and protactinium on iron and nickel sulfides; and cesium was coprecipitated with nickel ferrocyanide. The concentrated fission products, if the form of suspensions, were placed in long-term storage facilities.

The clarified solution was acidified with nitric acid and concentrated by evaporation. Simultaneous with the evaporation, acetic acid was distilled and recovered in a plate column sprayed with sodium hydroxide. The recovered sodium acetate was returned to the process.

The evaporator bottoms typically contained 1,100 to 1,150 g/L of sodium nitrate. Crystallization and recrystallization purified sodium nitrate if higher purity was required.

Mayak^[25, 26, 27] apparently swapped to an ion exchange process after the 1957 accident (Kyshtym disaster) and then to the PUREX process in 1982. Tomsk-7 was perhaps the last plant to swap to the Purex process. This change was made in 1983.^[24]

PLUTONIUM RESIDUE RECOVERY AND PLUTONIUM RECYCLE

United States

As the need for plutonium rose and fell with the political situation of the "Cold War" and the US began to recycle older weapons many new processes came online. Dry or pyrochemical processes^[36–50] were introduced to:

- A. Remove ingrown ²⁴¹Am from the decay of ²⁴¹Pu and thereby lower the radiation dose to weapons fabricators and the weapons handlers in the field.
- B. Try to keep the volume of radioactive waste generated to a manageable level.

Rocky Flats started using Molten Salt Extraction to remove ingrown ²⁴¹Am from recycled weapons. Other processes such as direct oxide reduction and electrorefining were introduced. These processes, however, generated spent chloride salt residues that contained large amounts of plutonium. In general, efficient processes to recover the plutonium from these and other residues were either never developed or were never installed. As a result, the US began to collect large volumes of residues from the 1960s to 1988. Several programs^[51–55] were initiated to recover these residues in the 1970s and 1980s (Interim Plutonium Residue Recovery Plan {IPRRP} and the Materials Management Executive Committee {MMEC}, etc.). New facilities were constructed at Rocky Flats (Building 371) and vast amounts of residues were shipped from Rocky Flats to Hanford, Los Alamos, and Savannah River in an attempt to bring the residue situation under control. New processes, such as the Scrub Alloy Process, that involved two sites instead of one were introduced. Although metric tons of plutonium was recovered from the mountain of residues and returned to the weapons system by these programs, they never brought the residue problem under control because were never adequately funded.

Plutonium was discarded based upon the economic of recovering the plutonium from the residues. In general, residues discarded to waste contained no more than a 0.5% of plutonium by weight (see Table 1) although some very difficult to recover materials were discarded at about 1.5 wt% plutonium. In 1990, the discard limits for two sites were raised to about 5 to 6 wt% Pu. In 1996, DOE/OSS issued a new set of guidelines^[57,58] which:

- Allows reclassification of low-grade materials based upon consideration of both plutonium concentration and the relative attractiveness for recovery
- Threshold level A represents the concentration at which safeguards can be terminated for various forms of low-grade plutonium bearing materials

• Threshold level B represents the concentrations at which physical protection measures can be reduced to measures equivalent with Category IV requirement if conditions in DOE Orders 5633.3 B are met.

These guidelines are given in Table 3.

TABLE 1. 1985 ECONOMIC DISCARD LIMIT STUDY

	Grams Pu per kg Bulk			
Residue Category	Site A	Site B	Site C	Site D
1. Graphite	0.6		9.8	0.8
2. Combustibles	1.2		2.9	1.4
3. Ash	5.4	10.3	15.6	5.6
4. Heels	5.9	14.6		2.2
5. Sand, Slag & Crucible	3.8	4.5	1.1	7.0
6. Insulation & Filters	4.8		3.1	2.8
7. Ceramics	6.8			1.2
8. Scrap Metal	0.8		2.9	1.0
9. Glass	1.5			2.9
10. Leaded Rubber	1.9		3.0	0.7
11. Sludge	3.9	15.2		5.0
12. Salts	16.0			5.0

TABLE 2. 1990 PLUTONIUM DISCARD LIMIT STUDY

	Grams Pu	Grams Pu per kg Bulk	
Residue Category	Site A	Site B	
A. Graphite	50	56	
B. Combustibles	51	59	
C. Ash	50	65	
D. Heels	42		
E. Sand, Slag & Crucible	11	41	
F. Insulation & Filters	134	54	
G. Ceramics	43	56	
H. Scrap Metal	51	65	
I. Glass		44	
J. Leaded Rubber	49	95	
K. Sludge	42		
L. Salts			
MSE		52	
—ER		48	
—DOR		53	

-12-

TABLE 3. ADDITIONAL ATTRACTIVENESS LEVEL E CRITERIA FOR SNM

		Threshold (wt%)	
CATEGORY	DESCRIPTION/FORM	A	В
Readily recoverable	SNM solutions and oxides: nitrate, caustic, chloride solutions, contaminated/impure oxides, metal fines and turnings, glovebox sweepings	0.1	N/A
Recoverable	SNM amenable to dissolution & subsequent separations: pyrochemical salts, chloride melt, hydroxide cake, floor sweepings, alumina, condensates, reduction residues, SS&C, MgO crucibles	0.1	0.2
Difficult to Recover	SNM in organic matrices or requiring pyrochemical separations, disassembly, and subsequent multiple recovery operations: HEPA filters, organic solutions, oils and sludges, graphite or carbon scrap, surface contaminated plastics, metal components, combustible rubber	0.2	1.0
Extremely Difficult to Recover	SNM bound in matrix or solid sintered or agglomerated refractory materials: SNM embedded in glass or plastic, high fired incinerator ash, special resins, salt sludges, raffinates and sulfides		2.0
Practically Unrecoverable			5.0

Improvements were also made to the PUREX process^[59-64] and many spent fuels that had been labeled as non-processable were processed. Processes were developed that allowed many different residues^[70-77] to be introduced into the solvent extraction streams of the PUREX process for purification and recovery. The plutonium was returned to the weapons system.

Russia

In general, Russia has always treated her plutonium as if it were a national treasure. By law, the Russian processing sites must recover plutonium from all residues in which the plutonium concentration is greater than 200 ppm. In general, nitric acid dissolution is used first; all residues not meeting the discard limit are then fused with NaOH (eight times their weight) at about 800°C. The fused residue is then subjected to nitric acid dissolution. Residues that do not meet the discard limit are fused and dissolved again and again until they meet the limit.

THE PLUTONIUM PROBLEM TODAY

The National Academy of Sciences (NAS) Committee on International Security and Arms Control^[78,79] has characterized the excess fissile materials as constituting a "clear and present danger" to national and international security.^[78,79] The NAS also stated that "The 'reactorgrade' Pu ... while it could be used to make nuclear bombs, it poses much smaller risks than separated Pu ... because of the mass, bulk, and intense radiation field of the spent fuel assemblies and because of the additional technical sophistication and resources required for chemical separation ... from the accompanying fission products and uranium."

The recommendation of the NAS^[78,79] was that options for long-term disposition of the excess plutonium should seek to meet the "spent fuel standard." The NAS definition of the "Spent fuel standard" is to "make the weapons-usable plutonium roughly as unattractive and as inaccessible for retrieval and weapons reuse as the residual and growing stockpile of plutonium in spent fuel from commercial reactors."^[78,79]

The NAS^[78,79] further stated, "Options for the disposition of WPu that leave it more accessible than the Pu in spent reactor fuel would mean that the WPu would continue to pose a unique safeguards problem indefinitely. Conversely, accepting substantial costs, complexities, risks, and delays in order to go beyond the "Spent Fuel Standard" (SNF) to make the WPu significantly less accessible for weapons use than the Pu in commercial spent fuel would not be justified unless the accessibility of the global stock of Pu in spent fuel were to be similarly reduced."

There are many steps in the manufacture of plutonium parts for nuclear weapons. [18,19] The sudden shutdowns of plants that did this work, including the Rocky Flats Plant in Colorado, the Hanford Site in Washington, the Savannah River Site in South Carolina, stranded 26 tonnes of plutonium in various intermediate steps. The plutonium is in a wide variety of forms, from plutonium dissolved in acid to rough pieces of metal to nearly finished weapons parts. Scraps of metal and chemicals that contain enough plutonium to be worth recovering were stored in drums and cans. Unknown amounts of plutonium have collected on the surfaces of ventilation ducts, air filters and gloveboxes.

P-8 SUMMIT ON NUCLEAR SAFETY AND SECURITY

Leaders^[80] of the P-8 nations ("G-7" countries of Canada, France, Germany, Great Britain, Italy, Japan, and the United States plus Russia) held the P-8 Summit on Nuclear Safety and Security in Moscow on April 19 – 20, 1996, to focus attention on the pressing international problem resulting from the large quantities of fissile materials becoming excess to military needs. At the summit, the leaders expressed their determination:

- 1. "To identify appropriate strategies for the management of fissile material designated as no longer required for defense purposes";
- 2. To ensure the "fissile material designated as no longer required for defense purposes will never again be used" in nuclear explosives;
- 3. To ensure that these materials "are stored and handled under physical protection, accounting and control measures that meet the highest international standards and that ensure effective non-proliferation controls",

-14- 4/15/99

- 4. That these materials should be placed under International Atomic Energy Agency safeguards "as soon as it is practical to do so",
- 5. "That effective management of this material will aim to reduce stocks of separated plutonium and highly-enriched uranium ... as soon as practical"; and
- 6. That the result of disposition of these materials should be that they are "transformed into spent fuel of other forms equally unusable for nuclear weapons." This last goal is called the "Spent Fuel Standard."

PRESENT SITUATION

With the end of the Cold War the Department of Energy underwent a profound shift^[81] in mission moving from a primarily weapons production mission to one of cleaning up the environmental legacies of the Cold War. This shift in the Department's mission from defense-related production to environmental cleanup of Cold War legacies has fundamentally altered the need for and management of nuclear materials. Previously each nuclear material had defined uses and processes by which the material was managed. Now, the change of mission has left large amounts of nuclear material without defined uses, and the management processes that so long governed these uses have not always been maintained or modified to reflect the new reality.

The Department of Energy has in place a system for planning and managing the use of nuclear materials that still have programmatic use. This has been a traditional role for the DOE, and the organizations exist to provide this management, responsibilities have been documented, and these programs are conducted on a continuing basis. For example, Defense Programs (DP) produced, processed, managed, stored, and distributed essentially all nuclear materials for the Department of Energy (DOE), and most materials were in active use by Defense Programs. This program was, and continues to be, responsible for the management and funding for their materials from use through disposition including, storage, stabilization, handling, characterization, packaging and shipping.

The Office of Nuclear Energy, Science and Technology (NE) maintained responsibility for:

- 1. Fuel fabrication for and operation of the Department's nuclear research and test reactors;
- 2. Production of Pu-238 from the irradiation and processing of Np-237 targets for radioisotope thermoelectric generators (RTG) for NASA missions;
- 3. The Departmental isotope production and distribution program; and
- 4. The disposition program for depleted uranium hexaflouride.

The Office of Fissile Materials Disposition (MD) has responsibility for policy development and evaluation of options for the disposition of most surplus (not needed for any program use) weapons-capable fissile materials (~50 metric tons of plutonium and ~174 metric tons of highly enriched uranium).

Additionally, the Office of Environmental Management (EM), originally created to disposition surplus facilities and to manage the Department's nuclear waste, has accepted custody of substantial quantities of non-waste, excess nuclear materials and is involved in the stabilization and storage of nuclear materials. EM owns most of the excess nuclear materials and many of the facilities with capability to provide stabilization, storage, and disposition.

-15- 4/15/99

Several Defense Nuclear Facilities Safety Board (DNFSB) Recommendations directly aimed at modifying the Department's management of its nuclear materials; generally in the case of DNFSB Recommendation 94-1 and specifically uranium-233 in the case of DNFSB Recommendation 97-1, have been assigned to EM to manage.

Plutonium is identified^[31] as weapon-grade, fuel-grade, or power reactor-grade based on the percentage of plutonium-240 that is contained in the plutonium. Weapon-grade plutonium contains less than 7 % 240 Pu. Fuel-grade plutonium contains from 7% to <19% 240 Pu. Power reactor-grade plutonium contains \geq 19% 240 Pu. In addition, DOE has defined weapons-usable plutonium^[82] to include all plutonium except plutonium present in spent nuclear fuel and plutonium containing greater than 10% 238 Pu. The present US inventory is composed of 85.0 tonnes of weapon-grade, 13.2 tonnes of fuel-grade, and 1.3 tonnes of power reactor-grade. Of the 85.0 tonnes of weapon-grade plutonium, 38.2 tonnes have been declared excess to national security needs.

Some of the plutonium declared excess consists of "pits" or triggers^[31,82] from dismantled weapons. This material meets the weapons specifications for impurities — it therefore, is relatively high purity material. With the possible exception of gallium, it should be straightforward to convert this material to plutonium oxide suitable for use in reactors as MOX fuel.

The remainder of the surplus plutonium^[18,19,31] comes from various sources including:

- A. Residues from weapons manufacture,
- B. Materials left in process when the weapons complex was shut down at the end of the Cold War,

-16-

- C. Experimental reactor fuels that were prepared but not irradiated, and
- D. Other miscellaneous materials.

A general categorization of these materials^[18,19,31] left "in the pipeline" is as follows:

- 1. 388,000 liters of plutonium solutions
- 2. 6,450 plutonium metal items
- 3. 8,405 plutonium oxide items, and
- 4. 33,288 plutonium residue items.

Full chemical characterization of most of these plutonium materials was never made. During the "Cold War" these materials were stored for recycle through a purification line and then back into the weapons stream. Therefore, only the data required to maintain accountability and to decide which processes would be necessary to purify these materials was maintained. To characterize and preparing these for use in MOX fuel would be very costly and result in considerable worker exposure to radiation.

DOE has directed the Lawrence Livermore National Laboratory^[83-89] to develop the processes to immobilize this material in a titanate-based ceramic form and then embed the ceramic forms in high level waste glass.^[90-125] The preferred site for this immobilization operation is the Savannah River Site near Aiken, South Carolina.^[87]

PRINCIPAL OBJECTIVES OF DOE PLUTONIUM-239 PROGRAMS

DOE has published^[126] its goals for the plutonium programs. These are:

- 1. Stabilize and package materials to meet 94-1 Implementation Plan Requirements
- 2. Consolidate materials to accelerate closure of RFETS and enable earlier closure of Hanford
- 3. Convert 50 tonnes of surplus plutonium into form meeting the spent nuclear fuel standard, and
- 4. Achieve other program (e.g., DP/NE) requirements.

PROPOSED ACTION BY THE UNITED STATES

In March 1995, the President of the United States declared 200 metric tons of fissile material surplus to national needs. These materials are in various compositions and forms. A long-term storage plan is needed to provide continued adequate control of these surplus materials and any that may be declared surplus in the future, from now through final disposition, as well as management and storage of nonsurplus fissile material that will not be subject to disposition. [126]

The initial quantities of fissile material were approximately 174 tonnes of high-enriched uranium and 38 tonnes of plutonium. The initial quantities have since undergone some exchanges as the Department of Defense (DOD) and the Department of Energy (DOE) has evaluated exact material requirements. The latest estimates for plutonium are given in Table 4. [119]

-17- 4/15/99

TABLE 4. COMPOSITION OF US SURPLUS PLUTONIUM BY FORM AND GRADE

Form	Weapon-Grade ¹	Fuel-Grade ²	Total
Metal ³	27.8	1.0	28.8
Oxide ⁴	3.1	1.2	4.3
Reactor Fuel ⁵	0.2	4.2	4.4
Irradiated Fuel ⁶	0.6	6.1	6.7
Other Forms ⁷	6.4	0.7	7.1
Totals	38.2	13.2	51.3

- 1. Weapon-grade plutonium contains less than 7% ²⁴⁰Pu.
- 2. Fuel-grade plutonium contains from 7% to <19% ²⁴⁰Pu.
- 3. Metal refers to plutonium in weapon components, ingots and buttons.
- 4. Oxide refers to plutonium oxide powder
- 5. Reactor fuel refers to fabricated mixed-oxide fuel and metal alloy fuel elements, pellets and mixed-oxide powder.
- 6. Irradiated fuel refers to mixed-oxide fuel and metal alloy fuel.
- 7. Other forms refer to enriched uranium/plutonium oxides and residual process materials from the fabrication of weapons components.

The US Department of Energy has proposed to take the following actions^[126, 127] for the US excess weapons-usable fissile materials:

- 1. Long-Term Storage provide a storage system (for up to 50 years) for Pu and HEU that meets the Stored Weapons Standard and applicable environmental, safety, and health standards while reducing storage and infrastructure
- 2. Disposition make surplus Pu and Pu that may be declared surplus in the future as inaccessible and unattractive for weapons use as the Spent Fuel Standards dictates, thereby providing evidence of irreversible disarmament and setting a model for proliferation resistance.

In the Programmatic Environmental Impact Statement and Record of Decision for the Storage and Disposition of Weapons-Usable Fissile Materials (January 14, 1997), DOE announced pursuit of two disposition technologies:

- 1) Irradiation of Pu as MOX fuel in existing reactors and
- 2) Immobilization of Pu into solid forms containing fission products as a radiation barrier.

DOE chose an immobilization approach that includes "use of the can-in-canister option... for a portion of the surplus, non-pit Pu material." In the can-in-canister approach, cans of ceramic forms containing Pu are encapsulated within canisters and surrounded by high level waste (HLW) glass. The primary isotope in the HLW glass will be ¹³⁷Cs; the design radiation rate is >100 Rad/hr at one meter, 30 years after fabrication, or >200 R/hr at fabrication. This is roughly equivalent to 40-year-old BWR fuel.

-18- 4/15/99

At present, the material to be immobilized exists in many forms and locations around the country, and is under the control of several DOE program offices. Prior to disposition, many of these materials will require stabilization in response to DNFSB Recommendation 94-1 and/or repackaging. It is assumed that the Materials Disposition Program (MD) within the Office of Fissile Materials Disposition will be receiving excess fissile materials packaged by the Offices of Defense Programs (DP), Environmental Management (EM), and Nuclear Energy (NE) facilities. It is assumed that the EM facilities will either prepare the plutonium as necessary to meet both nonproliferation concerns and transportation requirements for transferring the material to the Waste Isolation Pilot Plant (WIPP) or stabilize the material for transferring to MD. It is also assumed that DP and NE sites will package their excess materials as necessary and transfer them to MD.

Major objectives of the plutonium immobilization project^[119] are to:

- 1. Accommodate the largest possible range of plutonium feedstocks while minimizing feed characterization costs.
- 2. Minimize handling/processing of the material and therefore minimize the radiation dose to operators,
- 3. Minimize disruption to the EM (94-1) Stabilization Program, and
- 4. Minimize the final immobilization product variations and formulation space.

Once received, the planned immobilization facilities will have only limited capabilities to remove impurities prior to preparing the plutonium feedstocks for immobilization. Instead, blending will be done to levelize the impurities, the plutonium isotopics, and the uranium content.

The Center for Strategic and International Studies (CSIS) Senior Policy Panel on the Safe, Timely, and Effective Disposition of Surplus US and Russian Weapons-Grade Plutonium^[128] states that the "DOE decision to pursue a dual-track approach to plutonium disposition to be soundly conceived for several important reasons:

- 1. The pursuit by the United States of two avenues for achieving the spent-fuel standard will provide the nation with important insurance that it will have at least one credible route available for converting the US stockpile to the spent-fuel standard on a timely bases...
- 2. ... the pursuit of a MOX fuel irradiation approach as will as immobilization will send stronger and clearer assurances to Russia and others that the US plutonium-disposition program will, in fact be irreversible and impede any reuse of the surplus materials in nuclear weapons...
- 3. The use of a dual-track approach will also be essential to assure that the US disposition program receives the requisite support to succeed domestically and internationally...
- 4. The anticipated costs of the Russian and US disposition programs, while measured in the low billions of dollars, are modest when one considers the important security benefits that will be gained by achieving the spent-fuel standard...
- 5. From a policy perspective, the administration's dual-track decision is fully compatible with existing US nonproliferation policy because it will serve to reduce the global stocks of separated plutonium that are vulnerable to theft or diversion...

-19- 4/15/99

6. Finally, there also are ample grounds for concluding that the facilities that will be involved in the US program can be effectively safeguarded to discourage any theft, diversion, or misuse of the materials..."

To meet the DNFSB 94-1 stabilization requirements, Rocky Flats has requested and has been granted a variance from the safeguards termination limits setup by DOE/NN on July 22, 1996. This document states that recoverable items amenable to dissolution and subsequent separations such as pyrochemical salts, chloride melts, etc., are subject to safeguards terminations at 0.1-wt% plutonium. The variance requested by Rocky Flats and granted by DOE/NN was for all plutonium in containers less than 10-wt%, with blending of up to 20-wt% plutonium allowable so long as they were down blended to 10-wt% or less. This is a factor of 100 greater than is stated in the July 1966 document, and a factor of 500 greater than the discard limit used by the Russians.

Russia

The Russian Government views their excess plutonium as a national treasure to be used by the people for energy. They view any weapons-grade plutonium that will only be immobilized as being easier to reincorporate into weapons of existing design without testing than will plutonium that has been irradiated in reactors. In contrast to the United States, they do not view it as a waste and a liability. Therefore, the only option that is being seriously considered by the Russians is the MOX route. They however, would prefer the fast breeder route and the establishment of a plutonium economy. As a result, Russia is discarding only residues containing less than 0.02-wt% plutonium.

-20- 4/15/99

REFERENCES

- 1. Seaborg, Glenn T., *The Discovery of Plutonium in the Cyclotron*, Chapter 1 in **The Metal Plutonium**, A. S. Coffinberry and W. N. Miner, editors, The University of Chicago Press, Chicago, 1961.
- 2. Hoffman, Darleane C. [G.T. Seaborg Institute for Transactinium Science, Isotope Science, Lawrence Livermore National Laboratory, Livermore, CA (USA), 1996], *The transuranium elements: for neptunium and plutonium to element 112*, Report No. UCRL-JC-124718. CONF-9607139-3, Lawrence Livermore National Laboratory, Livermore, CA (USA).
- 3. Nuclear Successor States of the Soviet Union, Nuclear Weapons and Sensitive Export Status Report Number 4, The Monterey Institute of International Studies, Monterey, CA and The Carnegie Endowment for International Peace, Washington, DC and Moscow, May 1996.
- 4. Nash, Kenneth L. and Choppin, Gregory R., Separation's Chemistry for f Elements: Recent Developments and Historical Perspectives, Ninth Symposium on Separation Science and Technology for Energy Applications.
- 5. Fermi, E. (1934), Possible production of Elements of Atomic Number Higher than 92, Nature 133, 898.
- 6. Noddack, L. (1934), *Uber das Element 93*, **Angew. Chem. 47**, 653-655.
- 7. Hahn, O., Meitner, L., Strassmann, F., (1936), Elemente jenseits Uran, Berichte 69, 905-919.
- 8. Curie, L., Savitch, P. (1935), Sur la nature du radioelement de periode 3.5 heures forme sans l'uranium irradie par les neutrons, Compte Rendus 206, 1643-1644.
- 9. Hahn, O., Strassmann, F. (1939), *Uber den Nachweis und das Verhalten der bei der Bestrahlung des Uras mittels Neutronen entstehenden Erdakalimetalle*, **Naturwissenschaften** 27, 11-15.
- 10. Meitner, L. and Frisch O.R. (1939), Disintegration of Uranium by Neutrons: A New Type of Nuclear Reaction, Nature (letters) 143, 239-240.
- 11. Frisch O.R. (1939) Physical Evidence for the Division of Heavy Nuclei under Neutron Bombardment, **Nature (Letters)** 143, 276.
- 12. McMillan, E.M. and Abelson, P.H. (1940), *Radioactive Element 93*, **Phys. Rev. 57**, 1185-11 S6.
- 13. Seaborg, G.T., McMillan, E.M., Kennedy, J.W., and Wahl, A. C. (1946), *Radioactive Element 94 from Deuterons on Uranium*, **Phys. Rev**. 69, 366-367.

- 14. Seaborg, G.T., Wahl, A. C., and Kennedy, J.W., (1946), *Radioactive Element 94 from Deuterons on Uranium*, **Phys. Rev.** 69, 367.
- 15. Kennedy, J.W., Seaborg, G.T., Segré, E., and Wahl, A. C., (1946) Properties of 94 (239), **Phys. Rev.** 70, 555-556.
- 16. Seaborg, G.T., (1955), *Transuranium Elements: The Synthetic Actinides*, **Radiochimica Acta** 70/71, 69-90.
- 17. Seaborg, G.T., (1983), Forty Years of Plutonium chemistry: The Beginnings, Chapter 1 in Plutonium Chemistry, William T. Carnall and Gregory R. Choppin, Editors, ACS Symposium Series 216, American Chemical Society, Washington, DC.
- 18. The 1996 Baseline Environmental Management Report, Volume I, DOE/EM-0290, US Department of Energy, Office of Strategic Planning and Analysis, Washington, DC June 1996.
- 19. Closing the Circle on the Splitting of the Atom, US Department of Energy, Office of Environmental Management, January 1995.
- 20. Richard Rhodes, *The Making of the Atomic Bomb*, Simon and Schuster, New York, 1986.
- 21. Fritz Weigel, Joseph J. Katz and Glenn T. Seaborg, *Plutonium*, J. J. Katz, G. T. Seaborg, and L. R. Morss, editors, The Chemistry of the Actinide Elements, Second Edition, Volume 1, Chapman and Hall, Ltd., New York (1986).
- 22. Manson Benedict, Thomas H. Pigford, and Hans Wolfgang Levi, *Nuclear Chemical Engineering*, McGraw-Hill, Inc. San Francisco, 1981.
- 23. J. M. Cleveland, *The Chemistry of Plutonium*, American Nuclear Society, La Grange Park, IL, 1979.
- 24. Leonard W. Gray, F. Charles Gilbert, Fred E, Witmer, Roland E. Felt, Lester Harold Sullivan, Donald D. Wodrich, and Monte Lee Hyder, *Trip Report: Moscow and Tomsk, Russia, June 19 29, 1993*. Follow-up to the Tomsk-7 Accident, US Department of Energy, Office of the Assistant Secretary for Defense Programs, Washington, DC, September 1993.
- 25. D. J. Bradley, *Radioactive Waste Management in the Former USSR: Volume III*, PNL-8074, Pacific Northwest Laboratory, Richland Washington, June 1992.
- 26. D. J. Bradley and K. J. Schneider, *Radioactive Waste Management in the USSR: A review of Unclassified Sources*, 1963 1990, PNL-7182, Pacific Northwest Laboratory, Richland Washington, March 1990.
- 27. D. J. Bradley, Radioactive Waste Management in the USSR: A review of Unclassified Sources, Volume II, PNL-7645, Pacific Northwest Laboratory, Richland Washington, March 1991.

-22- 4/15/99

- 28. M. S. Gerber, Legend and Legacy: fifty Years of Defense Production at the Hanford Site, WHC-MR-0293, Revision 2, Westinghouse Hanford Company, Richland, Washington, September 1992.
- 29. Leslie R. Groves, *Now It Can Be Told The Story of the Manhattan Project*, Harper and Brothers publishers, New York, 1962.
- 30. Michael B. Stoff, Jonathan F. Fanton, and R. Hal Williams, *The Manhattan Project; A Documentary Introduction to the Atomic Age*, Temple University Press, Philadelphia, 1991.
- 31. Plutonium: *The First 50 Years; United States Plutonium Production, Acquisition, and Utilization from 1944 through 1994*, US Department of Energy, Washington, D.C. (U.S.A.). Report No. DOE/DP-0137. SuDoc. No. E 1.99:DE96009911 (1996).
- 32. Richard Rhodes, The Making of the Atomic Bomb, Simon and Schuster, New York, 1986.
- 33. Hanford Works, *Hanford Works Separations Section Operating Standards, Part I. Bismuth Phosphate and Isolation*, HW-17398, October 15, 1951, Report No. HW-17398. Hanford Works, Richland, WA (USA).
- 34. Warf, J. C., J. Amer. Chem. Soc. 71: 2187 (1949).
- 35. McKibben, J. M., Chemistry of the Purex Process, Radiochimica Acta, 36, 3 15 (1984).
- 36. Eldon L. Christensen, Leonard W. Gray, James D. Navratil, Wallace W. Schulz, *Present Status and Future Directions of Plutonium Process Chemistry*, Chapter 23 in **Plutonium Chemistry**, William T. Carnall and Gregory R. Choppin, Editors, ACS Symposium Series 216, American Chemical Society, Washington, DC.
- 37. Charles E. Baldwin and James D. Navratil, *Plutonium Process Chemistry at Rocky Flats*, Chapter 24 in **Plutonium Chemistry**, William T. Carnall and Gregory R. Choppin, Editors, ACS Symposium Series 216, American Chemical Society, Washington, DC.
- 38. Melvin Coops, James B. Knighton, and Lawrence J. Mullins, *Pyrochemical Processing of Plutonium*, Chapter 25 in **Plutonium Chemistry**, William T. Carnall and Gregory R. Choppin, Editors, ACS Symposium Series 216, American Chemical Society, Washington, DC.
- 39. D. C. Christensen and L. J. Mullins, *Plutonium Metal Production and Purification at Los Alamos*, Chapter 26 in **Plutonium Chemistry**, William T. Carnall and Gregory R. Choppin, Editors, ACS Symposium Series 216, American Chemical Society, Washington, DC.
- 40. Mullins, L. J.; Foxx, C. L., *Direct Reduction of* ²³⁸*PuO*₂ *to Metal*, US DOE Report LA-9073, Los Alamos National Laboratory, Los Alamos, N. M., January 1982.

-23-

4/15/99

- 41. Mullins, L. J.; Christensen, D. C.; Babcock, B. R., Fused Salt Processing of Impure Plutonium Dioxide to High-Purity Metal, US DOE Report LA-9154-MS, Los Alamos National Laboratory, Los Alamos, N. M., January 1982.
- 42. Baker, R. D., *Preparation of Plutonium Metal by the Bomb Method*, US AEC Report LA-473, Los Alamos Scientific Laboratory, Los Alamos, NM, 1946.
- 43. Mullins, L. J. Morgan, A. N., A Review of Operating Experience at Los Alamos Plutonium Electrorefining Facility, 1963-1977, US DOE Report LA-8943, Los Alamos National Laboratory, Los Alamos, NM, December 1981.
- 44. Christensen, E. L.; Maraman, W. J., *Plutonium Processing at the Los Alamos Scientific Laboratory*, US AEC Report LA-3542, Los Alamos Scientific Laboratory, Los Alamos, NM, April 1969.
- 45. Knighton, J. B.; Auge, R. C.; Berry, J. W.; Franchini, R. C., *Molten Salt Extraction of Americium from Molten Plutonium Metal*, US ERDA Report RFP-2365, Dow Chemical Co., Golden Colorado, March 12, 1976.
- 46. DeGrazio, R. P., A Prototype Hydriding Apparatus (Application for Plutonium Recovery), US ERDA Report RFP-2340, Dow Chemical Co., Golden Colorado, March 17, 1975.
- 47. Conner, W. V., *Process Studies on the Reduction of Plutonium Tetrafluoride to Metal*, US. AEC Report RFP-728, Dow Chemical Co., Golden Colorado, May 11, 1966.
- 48. Wade, Warren Z. and Wolf, T., *Preparation of Massive Plutonium Metal Directly from it's Oxides*, J. Nucl. Science and Technology, 6 (no. 7), 402-407 (1969).
- 49. Felt, R. E., A Pyrochemical Process for the Reduction of Plutonium Dioxide to Metal, US AEC Report ARH-1198, Atlantic Richfield Hanford Co., Richland, Washington, July 1969.
- 50. Long, J. L., Perry, C. C., The Molten Salt Extraction of Americium from Plutonium Metal, Nuclear Metal, 15, p.385 (1969).
- 51. Leonard W. Gray and J. H. Gray, Aqueous Recovery of Plutonium from Pyrochemical Processing Residues, Chapter 17 in Proceedings of the International Symposium on Actinide/Lanthanide Separations, G. R. Choppin, J. D. Navratil, and W. W. Schulz, eds., World Scientific, Singapore (1985).
- 52. J. H. Gray, L. W. Gray, and D. G. Karraker, *Recovery of Plutonium from Electrorefining Anode Heels at Savannah River*, US. Research and Development Report, DP-1732, March 1987.
- 53. Leonard W. Gray, *IPRP Flowsheet Development at SRL*, Invited paper for presentation at the Ninth Actinide Workshop, Richland, Washington, May 1985.

-24- 4/15/99

- 54. Leonard W. Gray and H. P. Holcomb, *Interfacing Solvent Extraction in the Recovery of Pyrochemical Residues at the Savannah River Plant*, DP-MS-86-167, Invited paper for presentation at the Sixth Pyrochemical Workshop, Los Alamos, New Mexico, October 1986.
- 55. Leonard W. Gray, D. A. Orth, and S. T. Augsburger, *Future Concepts of Pyrometallurgical Operations at the Savannah River Plant*, DP-MS-86-169, Invited paper for presentation at the Sixth Pyrochemical Workshop, Los Alamos, New Mexico, October 1986.
- 56. E. N. Moore, *Impact of Limits for the Reduction or Termination of Safeguards on Low-grade Residues*, WSRC-RP-96-489, Westinghouse Savannah River Company, September 1996.
- 57. Edward J. McCallum, Memo: "Additional Attractiveness Level E Criteria for Special Nuclear Materials" July 22, 1996.
- 58. E. B. Shelton, Experience with Processing Irradiated Fuel at the Savannah River Plant (1954 1976), DP-1467, Savannah River Laboratory, 1977.
- 59. W. P. Bebbington, *The Reprocessing of Nuclear Fuels, Scientific America*, December 1976, Vol. 235, No. 6.
- 60. J. M. McKibben, J. M. Starks, J. B. Brown, *The SRP Purex Plant 25 years of Successful Remote Operations*, Proceedings of the 27th Conference on Remote Systems Technology, American Nuclear Society National Meeting, 1979.
- 61. D. A. Orth, *Performance of 3% and 30% TBP Processes*, Proceedings International Solvent Extraction Conference, Harwell, England, 1965.
- 62. L. E. Bruns, *Plutonium-Uranium Partitioning by a Reflux Extraction Flowsheet*, Proceeding, International Solvent Extraction Conference 1971, Society of Chemical Industry, London 1971.
- 63. H. Pollock, *A Stable Diluent for Purex Process Extractants*, DP-294, Savannah River Laboratory, November 1958.
- 64. W. L. Poe, A. W. Joyce, R. I. Martens, ²³⁷Np and ²³⁸Pu Separations at the Savannah River Plant, I&EC Process Design and Development, Vol. 3, October, 1964.
- 65. L. W. Gray, Dissolution of Mixed Oxide and Mixed Alloy Reactor Fuels at the Savannah River Plant, US DOE Research and Development Report, DP-MS-82-92, presented at the National Meeting of the American Chemical Society, Seattle, Washington, March 20–25, 1983.
- 66. J. C. Chesna and L. W. Gray, *Dissolution of Uranium Fuel Tubes in Nitric Acid*, US DOE Research and Development Report, DP-1671, February 1984.

- 67. Leonard W. Gray, Reprocessing Fuel from the Southwest Experimental Fast Oxide Reactor at the Savannah River Plant, US DOE Research and Development Report, DP-1673, November 10985.
- 68. Leonard W. Gray, *Recent Solvent Extraction Experience at Savannah River*, DP-MS-86-59, Invited paper for presentation at the Tenth Actinide Workshop, Los Alamos, New Mexico, May 1986.
- 69. Leonard W. Gray, *Reprocessing of Nuclear Fuels at Savannah River Plant*, DP-MS-86-157, Invited paper for presentation at the S. C. Jr. Academy of Science, University of South Carolina—Aiken, October 1986. Presentation repeated at University of New Mexico (October 1986), New Mexico Institute of Mining and Technology (October 1986), Science Education Enrichment Day, University of South Carolina—Aiken (November 1986), and SOAR Program, A. L. Corbett Middle School (May 1987).
- 70. Leonard W. Gray, *Kinetics of the Ambient Temperature Dissolution of Plutonium Metal in Sulfamic Acid*, US DOE Research and Development Report, DP-1484, March 1978.
- 71. Leonard W. Gray and N. W. McGahee, *Recovery of Plutonium from Carbon Analysis Residues*, US DOE Research and Development Report, DP-1495, April 1978.
- 72. Leonard W. Gray, G. Ervin, and N. W. McGahee, *Recovery of Plutonium from Sodium Fluoride and Alumina Traps Loaded with Plutonium Tetrafluoride*, US DOE Research and Development Report, DP-1502, November 1978.
- 73. Leonard W. Gray, Removal of Sulfamic Acid from Plutonium Sulfamate-Sulfamic Acid Solutions, US. DOE Research and Development Report, DP-1503, October 1978.
- 74. Leonard W. Gray, Cation Exchange Process for Recovery of Plutonium from Laboratory Solutions Containing Chloride, US DOE Research and Development Report, DP-1508, October 1978.
- 75. Leonard W. Gray, Dissolution of Plutonium Metal in Sulfamic Acid at Elevated Temperatures, US DOE Research and Development Report, DP-1515, February 1979.
- 76. Leonard W. Gray, Rapid Dissolution of Plutonium Metal in Sulfamic Acid followed by conversion to a Nitric Acid Matrix, Nuclear Technology, 52, 66 (1981).
- 77. Leonard W. Gray and J. Radke, *Plutonium Scrap Recovery at Savannah River Plant*, Chapter 1 in <u>Actinide Recovery from Waste and Low Grade Sources</u>, J. D. Navratil and W. W. Shultz, eds., Harwood Academic Publishers, New York (1982).
- 78. National Academy of Sciences, Committee on International Security and Arms Control, *Management and Disposition of Excess Weapons Plutonium* (National Academy Press, Washington, D.C., 1994).
- 79. National Academy of Sciences, *Management and Disposition of Excess Weapons Plutonium* (National Academy Press, 1994), p. 34.

-26- 4/15/99

- 80. Joint United States/Russian Plutonium Disposition Study, September 1996.
- 81. Robert Price, Jay Bilyeu, and Michael Gates, Environmental Management Nuclear Materials Stewardship Program.
- 82. Office of Fissile Materials Disposition, Storage and Disposition of Weapons-Usable Fissile Materials Final Programmatic Environmental Impact Statement, US Department of Energy, Washington, DC, December 1996.
- 83. Office of Fissile Materials Disposition, Record of Decision for the Storage and Disposition of Weapons-Usable Fissile Materials Final Programmatic Environmental Impact Statement, US Department of Energy, Washington, DC, January 14, 1997.
- 84. Letter: Howard R. Canter, Office of Fissile Materials Disposition, DOE, to Steven G. Cochran, Lawrence Livermore National Laboratory, September 25, 1997.
- 85. DOE-MD Home Page Announcement Article, DOE Focuses on Ceramics for Immobilizing Surplus Plutonium.
- 86. William Danker, *Overview of US Plutonium Immobilization Program*, US Department of Energy, American Nuclear Society Third Topical Meeting **DOE Spent Nuclear Fuel and Fissile Materials Management**, Charleston, SC, September 8-11, 1998.
- 87. Letter: Steven G. Cochran, Lawrence Livermore National Laboratory, to Howard R. Canter, Office of Fissile Materials Disposition, DOE, August 27, 1997.
- 88. Letter: Matthew Bunn, Donald Langmuir, Ronald Loehman, David Stahl, Alan Williams Immobilization Down Select Peer Review Panel) to Stephen Cochran, Lawrence Livermore National Laboratory, August 21, 1997.
- 89. S. G. Cochran, T. A. Edmunds, L. M. MacLean, and T. H. Gould, Fissile Materials Disposition Program Final Immobilization Form Assessment and Recommendation, Lawrence Livermore National Laboratory, Livermore, CA, UCRL-ID-128705 (October 3, 1997).
- 90. Booth R. Myers, Guy A. Armantrout, Carol M. Jantzen, Adam Jostsons, J. Malvyn McKibben, Henry F. Shaw, Denis M. Strachan, and John D. Vienna, *Technical Evaluation Panel Summary Report: Ceramic and Glass Immobilization Options*, UCRL-ID-129315, Lawrence Livermore National Laboratory, January 1998.
- 91. Leonard W. Gray, Tehmau Kan, J. Malvyn McKibben, *Immobilization as A Route To Surplus Fissile Materials Disposition*, Proceedings of the Embedded Topical Meeting on DOE Spent Nuclear Fuel & Fissile Materials Management, Reno, Nevada, June 16 20, 1996, American Nuclear Society, Inc, LaGrange Park, Illinois, p 155ff.

- 92. L. W. Gray, T. Kan, W. G. Sutcliff, J. M. McKibben, and W. Danker, *Disposal of Surplus Fissile Materials via Immobilization*, Vol. XXIV, 413, Institute Of Nuclear Materials Management 36th Annual Meeting Proceedings, Palm Desert, CA, July 9 –12, 1995.
- 93. Leonard W. Gray, Bartley B. Ebbinghaus, Thomas A. Edmunds, Richard A Vankonynenburg, David C. Riley, Jay Spingarn, *Materials Disposition Acceptance Specifications for the Plutonium Immobilization Project*, PIP-98-047, (August 1998), Lawrence Livermore National Laboratory, Livermore, CA.
- 94. Leonard W. Gray, Mark Bronson, Bartly Ebbinghaus, and Guy Armantrout, *Dealing with a Dangerous surplus from the Cold War*, **Science and Technology Review**, Lawrence Livermore National Laboratory, Livermore CA, April 1997.
- 95. Bart Ebbinghaus, Tom Edmunds, Leonard W. Gray, and David Riley, *Preliminary Feed Materials Requirements and Blending Strategy*, PIP-98-008, January 16, 1998, Lawrence Livermore National Laboratory, Livermore, CA.
- 96. Sommer Gentry, Thomas Edmunds, and Leonard W. Gray, *Blending Strategy and Assumptions*, UCRL-ID-132206, PIP-99-013, (February 1999), Lawrence Livermore National Laboratory, Livermore, CA.
- 97. L. W. Gray and T. Kan, *Safety Aspects with Regards to Plutonium Vitrification Techniques*, Proceedings of the NATO Conference on "Disposal of Weapons Plutonium, St. Petersburg, Russia, May 14-17, 1995.
- 98. Leonard W. Gray Tehmau Kan, Leslie J. Jardine, Lawrence Livermore National Laboratory, Livermore, CA; J. Malvyn McKibben, Westinghouse Savannah River Company, Aiken, SC; William Danker, United States Department Of Energy, Washington, DC, *Immobilization as a Route to Surplus Fissile Materials Disposition*, published in the Proceedings of the ASME's Fifth International Conference on Radioactive Waste Management and Environmental Remediation, September 3–7, 1995, Berlin, Germany.
- 99. Leonard W. Gray and Jeff Kass, *Plutonium Immobilization* A Review of the Technology and R&D Programs, Presented to the Senior Technical Review Group, U. S. Department of Energy, February 1, 1996, Alexander, VA.
- 100. L. W. Gray, et. al, Fissile Materials Disposition Program, PEIS Data Call Input Report: Ceramics Immobilization Facility with Radionuclides, UCRL-ID-122665, Lawrence Livermore National Laboratory, Livermore, CA, February 9, 1996.
- 101. L. W. Gray, et al., Screening of Alternate Immobilization Candidates for Disposition of Surplus fissile Materials, UCRL-ID-118819, Lawrence Livermore National Laboratory, Livermore, CA, February 9, 1996.

- 102. T. Kan and L. W. Gray, *Status of Immobilization of Surplus Fissile Materials*, International Symposium on the Environmental Issues and Waste Management Technologies in Ceramics and Nuclear Industries, American Ceramic Society, April 15 17, 1996.
- 103. L. W. Gray, *The Plutonium Immobilization Option*, Third International Policy Forum: Management and Disposition of Nuclear Weapons Materials, March 19-22, 1996 Lansdown, VA.
- 104. L. W. Gray, T. Kan, H. Shaw, and G. Armantrout, *Immobilization Needs and Technology Programs*, Plutonium Stabilization & Immobilization Workshop, December 12-14, 1995, Washington DC, US Department of Energy, March 1996.
- L. W. Gray, T. Kan, W. G. Sutcliff, and J. M. McKibben, W. Danker, Disposition of Surplus Fissile Materials via Immobilization, Vol. XXIV, 413, Institute of Nuclear Materials Management 36th Annual Meeting Proceedings, Palm Desert, CA, July 9-12, 1995.
- 106. L. W. Gray, T. Kan. J. M. McKibben, *Immobilization As A Route To Surplus Fissile Materials Disposition*, **Proceedings of the ANS Imbedded Conference on Pu Disposition**, **Reno**, **NV**, June 1996.
- 107. L. W. Gray, *Immobilization of Surplus Weapons Plutonium*, Keynote Presentation for the "International Symposium on Glass and Ceramic Wasteforms, including Spent Fuel, for Radioactive Waste Management," The second International Meeting of the Pacific Rim Ceramic Societies, July 15-17, 1996, Cairns, Australia.
- 108. L.W. Gray, Overview: Proliferation Issues Surrounding Immobilization Of Surplus Fissile Materials, ANS Conference, Washington, DC, 1996.
- 109. L. W. Gray, G. A. Armantrout, H. Shaw T. Kan, *Plutonium Disposition Via Ceramic or Vitrification Immobilization*, Presented at the AIChE Meting, Houston, TX, March 9-13, 1997.
- 110. L. W. Gray, "The Next Nuclear Challenge—How do we Dispose of the Excess Nuclear Materials?", Presented at the Meeting of the Am. Chem. Soc., San Francisco, CA, April, 14 18, 1997.
- Bartley B. Ebbinghaus, Connie Cicero-Herman, Leonard Gray, Henry Shaw, *Plutonium Immobilization Project Baseline Formulation*, UCRL-ID-133089, PIP-99-012, Lawrence Livermore National Laboratory, February 1999.
- 112. Leonard W. Gray and Thomas H. Gould, *Immobilization Technology Down Selection:*Radiation Barrier Approach, UCRL-ID-127320, Lawrence Livermore National Laboratory (May 23, 1997).

-29-

- Thomas Gould, Booth Myers, Leonard Gray, Thomas Edmunds, Evaluation of Candidate Glass and Ceramic Forms for Immobilization of Surplus Plutonium, American Nuclear Society Third Topical Meeting DOE Spent Nuclear Fuel and Fissile Materials Management, Charleston, SC, September 8-11, 1998.
- DiSabatino, R. Jones, T. Kan, D. Loftus, P. Maddux, B. Marais, T. Severynse, *Design and Performance of a Plutonium Immobilization Production Plant*, American Nuclear Society Third Topical Meeting, **DOE Spent Nuclear Fuel and Fissile Materials**Management, Charleston, SC, September 8-11, 1998.
- William Brummond, Guy Armantrout, Paul Maddux, J. Vienna, *Ceramic Process Equipment for the Immobilization of Plutonium*, American Nuclear Society Third Topical Meeting **DOE Spent Nuclear Fuel and Fissile Materials Management**, Charleston, SC, September 8-11, 1998.
- 116. Leonard W. Gray, J. Malvyn McKibben, An Analysis Of Plutonium Immobilization Versus The "Spent Fuel Standard", American Nuclear Society Third Topical Meeting **DOE Spent Nuclear Fuel and Fissile Materials Management,** Charleston, SC, September 8-11, 1998.
- 117. Sommer Gentry, Thomas Edmunds, and Leonard W. Gray, *Blending Strategy and Assumptions*, UCRL-ID-132206, PIP-99-013, (February 1999), Lawrence Livermore National Laboratory, Livermore, CA.
- 118. Leonard W. Gray, Thomas A. Edmunds, Bartley B. Ebbinghaus, Sommer Gentry, and Richard A. Vankonynenburg, David C. Riley, and Jay Spingarn, *The Blending Strategy For The Plutonium Immobilization Program*, **Waste Management 99**, Tucson, AZ, February 28 March 4, 1999.
- 119. Guy Armantrout, Leonard W. Gray and Booth Myers, Thomas Gould, *Plutonium Immobilization Form Evaluation*, **Waste Management 98**, Tucson, AZ, March 1-5, 1999.
- 120. M. Bronson, J. Diaz, T.A. Edmunds, L.W. Gray, D.C. Riley, T.L. Rising, Immobilization Feed Stocks and Their Preparation for Immobilization, Waste Management 98, Tucson, AZ, March 1-5, 1999.
- 121. B. Ebbinghaus, T. Edmunds, L. Gray, D. Riley, T. L. Rising, Surplus Plutonium Immobilization Feed Materials Requirements and Blending Strategies, Waste Management 98, Tucson, AZ,—March 1-5, 1999.
- 122. B. Ebbinghaus, R. V. Konynenburg, H. Shaw, P. Curtis, W. Close, R. Burns, LLNL; E. Vance, M. Stewart, J. Allender, T. Rankin, J. Congdon, *Ceramic Formulation for the Immobilization of Plutonium*, **Waste Management 98**, Tucson, AZ, March 1-5, 1999.
- 123. W. Brummond, G. Armantrout, Ceramic Process Equipment for the Immobilization of Plutonium, Waste Management 98, Tucson, AZ, March 1-5, 1999.

-30- 4/15/99

- D. M. Strachan, A.J. Bakel, E. C. Buck, D. B. Chamberlain, J. A. Fortner, C. J. Mertz, S.F. Wolf, W.L. Bourcier, B. B. Ebbinghaus, H. F. Shaw, R. A. Van Konynenburg, B. P. McGrail, J. D. Vienna, J.C. Marra, D.K. Peeler, *The Characterization and Testing of Candidate Immobilization Forms for the Disposal of Plutonium*, Waste Management 98, Tucson, AZ, March 1-5, 1999.
- Thomas P. Grumbly, *The US Decision on the Disposition of Excess Weapons Plutonium*, Opening Keynote Address to the 4th **International Policy Forum: The Management and Disposition of Nuclear Weapons Materials**, February 11 –14, 1997, Lansdown, Virginia, Exchange/Monitor Publications & Forums, Lake Bluff, IL.
- 126. G. Klipa, Nuclear Materials Stewardship Program, *Materials Management Plan for Plutonium*–239, October 30, 1998.
- 127. John Taylor, Chair; Senator Pete V. Domenici and Representative Lindsey Graham, Project Cochairs; et al., *Disposing of Weapons-Grade Plutonium*; A Consensus Report of the CSIS Senior Policy Panel on the Safe, Timely, and Effective Disposition of Surplus US and Russian Weapons-grade Plutonium, March 1998.
- Office of Environmental Management, Linking Legacies, Connecting the Cold War Nuclear Weapons Production Processes to their Environmental Consequences, Office of Strategic Planning and Analysis (EM-24), US Department of Energy, Washington, DC, January 1997.
- 129. Russian Federation Nuclear Center, Russia's Nuclear and Thermonuclear Weapons, Russian Federation Nuclear Center, Sarov (Arzamas-16), Nishni Novgorod Region, Russia.
- 130. Thomas B. Cochran, William M. Arkin, Robert A. Norria, and Jeffrey I. Sands, *Nuclear Weapons Databook—Volume IV Soviet Nuclear Weapons*, Harper & Ron Publishers, New York, 1989.
- 131. The American Experience, Race for the Superbomb, PBS Online.
- 132. R. H. Condit, *Plutonium: An Introduction*, UCRL-JC-115357, Lawrence Livermore National Laboratory, Livermore, CA, October 1993.

-31-

-32-

APPENDIX

Timeline of Some Important Events for Plutonium

1781: Herschel, an English astronomer discovered the 7th planet Uranus. He first proposed to name it George, after King George III, but was talked out of it. (Ref. 132)

1789: Klaproth discovered a new element and named it after the newly discovered planet. Uranium was later identified as being the element with atomic number 92. (Ref. 132)

1846: Johann Galle of the Berlin Observatory discovered the 8th planet, Neptune. It was named after Neptune, the bearded sea god of the Romans.

November 8, 1895: German physicist Wilhelm Röntgen discovers x-rays. (Ref. 20)

1896: Henri Becquerel discovers radiation. (Refs. 19 & 132)

1905: Albert Einstein publishes his theory of relativity which included the equation, $E = mc^2$, which says that a little mass can be converted into a lot of energy. (Ref. 132)

1914: In the novel "The World Set Free," H. G. Wells predicted atomic bombs and atomic war. (Ref. 20)

1925: Swedish and German scientist publish estimates of "tolerance doses," the amount of radiation a person was thought to absorb without harm. Based on the amount of radiation that would burn skin, the tolerance dose was initially estimated to be the equivalent of about 156 rem per year. (Ref. 19)

1930: Tom Baugh in the US (Professor of Earth Science and Astronomy at New Mexico State University) discovered the 9th planet, Pluto. Tombaugh picked the name "Pluto" for the 9th planet, because it was cold and far from the sum. In Roman mythology Pluto was the brother of Neptune and god of the underworld. (Ref. 132)

1931: The tolerance dose is standardized at 6 rem per month (72 rem per year). (Ref. 19)

1932: Sir James Chadwick in England discovered the neutron. (Ref. 132)

1933: German scientist A. Mueller studies the genetic effects of radiation on fruit flies. He learned that radiation caused genetic mutations. (Ref. 19)

1934: The International Commission on X-Ray and Radium Protection publishes first international radiation safety standards based on measurements of damage to human tissue in Zurich. Workers are allowed up to 0.1 rem per day (30 rem per year). (Ref. 19)

-33- 4/15/99

- **1934:** E. Fermi, E. Amaldi, O. D'Agostino, F. Rasetti, and E. Segré discover that the neutron irradiation of uranium produced a number of radioactive substances. On the basis of their chemical work, Fermi and his co-workers concluded that these substances were transuranium elements:
 - Because they decayed by beta emission, a process that might result in elements of higher atomic number than uranium, and
 - Because they differed in chemical properties from all known elements in the region of uranium.

(Ref. 30)

December 1938: Two German physicists, Otto Hahn and Fritz Strassman discover the process of fission. In their work leading to the discovery of fission, they found that at least some of the radioactive substances produced by the bombardment of uranium with neutrons were isotopes of barium and lighter elements. Most of the radioactivities previously assigned by Fermi to the transuranium elements were found later to be due to fission products. (Refs. 30 & 128)

January 1939: Lise Meitner explained that the uranium atom could be split. Physical scientists realized that nuclear energy, derived from splitting the atom, might be used either to generate power or to create super-weapons. (Ref. 30)

1939: Word reaches Einstein through Hungarian-born physicist Leo Szilard that the Nazis had banned the export of uranium from Germany and German-occupied territory, including the rich Joachimsthal mines in Czechoslovakia. Einstein and Szilard concluded that the Germans were making a weapon of unprecedented force. (Ref. 30)

1939: Nobel Prize-winning physicist Niels Bohr, Danish physicist, had argued that building an atomic bomb "can never be done unless you turn the United States into one huge factory." Years later, he told his colleague Edward Teller, "I told you it couldn't be done without turning the whole country into a factory. You have done just that." (Ref. 19)

August 2, 1939: At the urging of the President's science adviser, Alexander Sachs, Einstein and Szilard composed a letter to President Roosevelt, though only Einstein signed the letter because he alone had sufficient status to attract presidential attention. "It is conceivable," the physicists warned the President, "... that extremely powerful bombs of a new type may thus be constructed." They beseeched Roosevelt to counter the German threat with an American atomic-bomb project. Roosevelt established a joint Army-Navy "Uranium Committee" to further study the question. (Ref. 30)

August 23, 1939: German-Russian non-aggression pact signed. (Ref. 30)

August 24, 1939: British-Polish mutual assistance pact signed. (Ref. 30)

September 1, 1939: Germany invades Poland setting off World War II. (Refs. 20, 30 & 128)

October 21, 1939: First meeting of the President's advisory committee on uranium. (Ref. 30)

-34- 4/15/99

November 1939: The "Uranium Committee" recommended that the military begin funding fission chain reaction research, already being conducted at several American universities. (Ref. 128)

June 1940: National Defense Research Committee was created under the chairmanship of Dr. Vannevar Bush. The Uranium Committee was made one of its subcommittees. (Ref. 30)

June 22, 1940: France surrenders to Germany. (Ref. 30)

Summer – Fall 1940: Battle of Britain. (Ref. 30)

September 2, 1940: American destroyers exchanged for British bases. (Ref. 30)

September 27, 1940: Germany, Italy, and Japan sign Tripartite Pact creating the Triple Alliance. (Ref. 30)

1940: Neptunium was the first of the transuranium elements to be discovered. Edwin M. McMillan and Philip H. Abelson produced it by bombarding uranium with neutrons. (Ref. 17)

February 23, 1941: Discovery of plutonium, an artificially produced fissile element, by a research group headed by Glenn T. Seaborg at the University of California in Berkeley. The first synthesized plutonium was ²³⁸Pu produced by bombardment of uranium with 16-Mev deuterons from the University of California 60-inch cyclotron and identified on February 23, 1941. Plutonium-238 was produced shortly thereafter by the ²³⁸U capture of neutrons produced by (d,n) reactions in the cyclotron. (Refs. 1, 17 & 132)

March 1941: Research group headed by Glenn T. Seaborg at the University of California – Berkeley produced the first submicroscopic amounts of plutonium-239. Naming element 94 after the planet Pluto was a natural choice, says Seaborg, because the previous two elements in the periodic table had been named "uranium" and "neptunium," after Uranus and Neptune. The association of the name, "Pluto", with the underworld has transferred somewhat to plutonium; there are those who like to call it the element of death. Plutonium was the first man-made element to be produced on an industrial scale. (Refs. 1, 17, 28, 128 & 132)

March 11, 1941: United States Congress approves lend-lease assistance to aid Allies. (Ref. 30)

April 13, 1941: Russian-Japanese five-year neutrality pact signed. (Ref. 30)

June 1941: The National Defense Research Committee re-formed into an advisory board to the Office of Scientific Research and Development and the S-1 Committee replaced the "Uranium Committee." (Ref. 128)

June 22, 1941: Germany invades Russia. (Refs. 30 & 130)

September 1941: Enrico Fermi suggests to Edward Teller that an atomic bomb might heat deuterium sufficiently to kindle a full-scale thermonuclear reaction. (Ref. 131)

-35- 4/15/99

September 1941: The cubical lattice that the Columbia football squad stacked in Schermerhorn Hall extrapolated to a disappointing first k of 0.87. The cans, which contained the uranium oxide, were made of iron, and iron absorbs neutrons. So the cans had to go. (Ref. 30)

December 7, 1941: Japan attacks Pearl Harbor and brings America into WWII. (Refs. 20, 28, 30 & 128)

December 8, 1941: United States declares war on Japan. (Ref. 30)

December 11, 1941: Germany declares war on United States. (Ref. 30)

January 1942: President Roosevelt approved the development of the atomic bomb. (Ref. 128)

April 20, 1942: Glenn Seaborg takes on the responsibility of the "94 chemistry group" at the Metallurgical Laboratory on the University of Chicago campus. (Refs. 1 & 17)

May 6, 1942: The Philippines falls to the Japanese. (Ref. 30)

June 17, 1942: Bush provides President Roosevelt with a report stating it was possible to make a nuclear weapon that could be employed in combat and it might be ready in time to influence that outcome of WWII. (Refs. 29 & 30)

June 18, 1942: Manhattan Engineer District (MED) was formed within the Army Corps of Engineers. The quest for nuclear explosives was inspired by the fear that Hitler's Germany might develop them first. This epic, top-secret engineering and industrial venture in the United States during World War II was termed the "Manhattan Project." (Refs 28 & 128)

Mid-1942: The United States Army Corps of Engineers began construction at Los Alamos. (Ref. 128)

Mid-1942: Government support resulted in research becoming concentrated at Columbia University in New York, the University of California in Berkeley, and the University of Chicago Metallurgical Laboratory. (Ref. 128)

August 20, 1942: Cunningham and Werner isolate approximately 1 μg of a pure plutonium compound from 90 kg of uranyl nitrate, which had been bombarded with cyclotron neutrons. (Refs. 1 & 17)

1942: Carrier precipitation test carried out proves effective in separating plutonium from uranium and fission products. (Refs. 1 & 17)

1942: Manhattan Project begins acquisition of uranium. Uranium was purchased from the Belgian Congo and Canada. (Ref. 128)

September 10, 1942: The first weighing of a plutonium compound carried out by Cunningham $-2.77 \mu g$ of plutonium dioxide. (Refs. 1, 17 & 20)

September 17, 1942: Brigadier General Leslie R. Groves named to head the MED. (Refs. 28, 29, 30 & 128)

November 1942: E. I. Du Pont de Nemours & Co. signed as prime contractor to construct and operate the plutonium production plant. For purely legal reasons, provision was made for a fee of one dollar. (Refs. 28, 29 & 128)

November 1942: Construction began at Los Alamos National Laboratory, Albuquerque, New Mexico. (Refs. 29 & 128)

November 16, 1942: Construction of Fermi's pile, CP-1, began in a double squash court under the west stands of Stagg Field. (Refs. 29 & 128)

December 1, 1942: The 57th layer of CP-1 was completed. The pile now contained 771,000 pounds of graphite, 80,590 pounds of uranium oxide, and 12,400 pounds of uranium metal. It cost about \$1 million to produce and build. Its only visible moving parts were its various control rods. CP-1 was simple and entirely a physics experiment designed to prove the chain reaction, unshielded and uncooled, and intended to run no hotter than half a watt. (Ref. 128)

December 2, 1942: Enrico Fermi and Leo Szilard obtained the first controlled, self-sustaining nuclear reaction. Fermi built three piles at Chicago. (Refs. 19, 20, 30, 128 & 132))

December 1942: Decision made to shift the plutonium production plant from Tennessee to another location. Du Pont had added it weight to the contention that plutonium production should take place far from the populated East and Midwest. (Refs. 28 & 29)

December 14, 1942: The Corps of Engineers and Du Pont developed the criteria for the plutonium production site. (Ref. 28)

December 1942 – January 1943: Dusty tract of land lying between the towns of White Bluffs, Hanford, and Richland, Washington chosen as the site of the first plutonium production plant. (Ref. 28)

January 14 – 23, 1943: Casablanca Conference: Franklin D. Roosevelt and Sir Winston Churchill announce policy of "unconditional surrender." (Ref. 30)

January 16, 1943: Hanford approved as the site of the first plutonium production plant. (Ref. 28)

February 8, 1943: Secretary of War's formal directive was issued, authorizing acquisition of the necessary land at the Hanford Site. The War Department original name was the "Gable Project." (Ref. 28)

March 1943: Rebuilding of CP-1 with modification as CP-2 was completed at the Palos Folrest Preserve outside Chicago. (Ref. 128)

-37- 4/15/99

March 1943: Groundbreaking for the Hanford Site. The Army Corps of engineers and the prime contractor E. I. Du Pont de Nemours & Co. officially named the site the "Hanford Engineer Works." (Ref. 28)

March 1943: Scientists and technicians began arriving at Los Alamos National Laboratory. (Ref. 128)

May 13, 1943: Surrender of Axis forces in North Africa. (Ref. 30)

June 1943: Construction began on B-Reactor at Hanford. (Ref. 128)

August 14 – 24, 1943: Quebec Conference: Roosevelt and Churchill negotiate Quebec Agreement. Despite their early contributions to the research, the British won access to the Manhattan Project only after vigorous protest. Their success, formalized in the Quebec Agreement, was the result of a geologic fact and a diplomatic pledge. The British Empire contained the largest natural reserves of fissionable material in the world, so their cooperation was essential. On top of that, they pledged not to use atomic secrets for commercial advantage after the war. (Ref. 30)

September 8, 1943: Surrender of Italy. (Ref. 30)

September 18, 1943: Roosevelt and Churchill meet at Hyde Park and agree on atomic policy. (Ref. 30)

Late 1943: The Calutrons at Oak Ridge become operational. Electromagnetic enrichment of uranium was done in a device called a "Calutron," a modification of an early cyclotron. The electromagnetic enrichment plant produced the first gram quantities of HEU in 1944. Two stages of electromagnetic "Calutrons" at the Y-12 Plant were used to produce all of the HEU for "Little Boy," the atomic bomb detonated over Hiroshima, Japan. Y-12 featured nine first-stage "alpha" racetracks and four second-stage "beta" racetracks. Uranium chloride was used for this purpose. (Ref. 128)

1943: Stalin initiated a small-scale atomic bomb project under the scientific direction of Igor Vasil'evich Kurchatov. (Ref. 130)

November 4, 1943: The air-cooled, pilot-scale reactor at Oak Ridge went critical at five o'clock in the morning. The pile, which was designated X-10, was a graphite cube twenty-four feet on a side drilled with 1,248 channels that could be loaded with canned uranium-metal slugs and through which large fans blew cooling air. The channels extended for loading through seven feet of high-density concrete that composed the pile face; at the back they opened onto a subterranean pool like the pools planned for Hanford into which irradiated slugs could be pushed to shield them until they lost their more intense short-term radioactivity. (Ref. 128)

November 22 – 26, 1943: Cairo Conference—Roosevelt, Churchill, and Chiang Kai-shek meet to discuss postwar Far Eastern policy. (Ref. 30)

-38- 4/15/99

November 28 – December 7, 1943: Teheran conference: First wartime meeting of "Big Three" (Roosevelt, Churchill, and Joseph Stalin), who agree on Anglo-American invasion of Western Europe and on postwar international security organization. (Ref. 30)

December 19, 1943: The bismuth phosphate process for extracting plutonium from irradiated uranium was demonstrated in a pilot plant alongside the Oak Ridge X-10 Reactor. This Hanford prototype was designed to separate one-tenth gram of plutonium for ton of reactor output. The first uranium from the X-10 Reactor was dissolved. (Ref. 128)

1944: United States Navy built a pilot scale thermal diffusion plant at the Philadelphia Navy Yard. The plant used concentric hot and cold pipes to provide the temperature differences needed to separate uranium isotopes. (In the presence of a temperature difference, the lighter uranium-235 isotope will diffuse toward a hot area faster than the heavier uranium-238 isotope.) (Ref. 128)

February 1944: X-10 began production of several grams of plutonium per month. (Ref. 128)

March 1944: Hanford's "triple dip" slug coating and canning process for bonding uranium slugs to the cans began operation. (Ref. 128)

May 1944: A heavy-water reactor, CP-3, started-up. (Ref. 128)

June 6, 1944: Allied invasion of Normandy, France (D Day). The invasion of Europe across the English Channel involved an initial force of 156,000 British, Canadian, and American soldiers supported by 1.200 warships, 1,5000 tanks, and 12,000 aircraft. Dwight David Eisenhower, the Supreme allied Commander, called it "The greatest amphibious assault ever attempted." By the end of the week, the Allies had secured the invasion beaches and begun advancing inland with a force bolstered now to 326,000 men. "The way home," Eisenhower instructed his armies, "is via Berlin." (Ref. 30)

June 8, 1944: Du Pont engineers in Wilmington, Delaware chose the bismuth phosphate for the full-scale plant. (Ref. 128)

September 26, 27 1944: The world's first plutonium production reactor, Hanford's B Reactor, starts up. Tuesday evening, September 26, 1944, the largest atomic pile yet assembled on earth was ready. It had reached dry criticality — the smaller loading at which it would have gone critical without cooling water if its operators had not restrained it with control rods — the previous Friday. Now the Columbia River circulated through 1,500 loaded aluminum tubes. With Fermi and the du Pont brass present the operators withdrew the control rods. Gradually gauges showed the cooling water warmed, flowing in at 50°F and out at 140°F. The pile went critical a few minutes past midnight.

-39- 4/15/99

If Du Pont had built the Hanford production reactors to Eugene Wigner's original specifications all three piles would have required complete rebuilding. Fortunately, Princeton theoretician John A. Wheeler had fretted about fission-product poisoning. After the massive wooden shield blocks that formed the front and rear faces of the piles had been pressed a year previously, Wheeler advise Du Pont to increase the count of uranium channels for a safety margin. Wigner's 1,500 channels were arranged cylindrically; the corners of the cubical graphite stacks could accommodate another 504. That necessitated drilling out the shield blocks, which delayed construction and added millions to the cost. Du Pont has accepted the delay and drilled the extra channels. They were in place now when they were needed; although not yet connected to the water supply.

Built in less than a year, the B Reactor operated from 1944 to 1946 and then from 1948 to 1968. B reactor had a design power level of 250 MWt. It was graphite moderated; light water cooled reactor with a single-pass cooling system. Plutonium created within B reactor fueled the first atomic explosion in the Alamogordo desert on July 16, 1945, and it formed the core of the bomb that exploded over Nagasaki on August 9, 1945. (Refs. 20, 29 & 31)

September 27, 1944: The pile in B Reactor was steadily decreasing with time. Control rods were withdrawn continuously from the pile to hold it a 100 megawatts. Early Wednesday evening B pile died. (Ref. 128)

September 28, 1944: Early Thursday morning B pile came back to life. By 7 am it was running well above critical again. But twelve hours later it began another decline. (Ref. 128)

1944: Y-12 at Oak Ridge began converting UO₃ to UCl₄ feed for the calutrons. (Ref. 128)

November 1944: The Calutron (California University Cyclotron) isotope separators at Y-12 (Oak Ridge) for electromagnetic separation of ²³⁵U become operational. These units produced the first kilogram quantities of highly enriched uranium in 1944. Uranium enrichment by Calutrons was discontinued in 1946. (Refs. 20 & 29)

November 1944: Uranium ingots began arriving at Hanford. (Ref. 128)

December 16 – 26, 1944: Battle of the Bulge. (Ref. 30)

December 17, 1944: D-Reactor at Hanford went critical with a full 2,004-tube loading. It had a design power level of 250 MWt. It was graphite moderated; light water cooled reactor with a single-pass cooling system. (Refs. 31 & 128)

-40-

December 26, 1944: T-Plant at Hanford, the world's first, large-scale plutonium separations facility, processes its first batch of irradiated fuel rods. The chemical process was a batch operation that used repeated dissolution, precipitation and centrifugation to separate plutonium from uranium and associated fission products. Separation was achieved by varying the plutonium valent state from +4 to +6. Plutonium remained in solution as the +4 but precipitated as the +6. This was the bismuth-phosphate process. The final Hanford product throughout WWII was a wet plutonium nitrate paste that was shipped to Los Alamos for conversion to metal. Due to the short cooling time of the fuel, significant quantities of iodine-131 (half-life of 8 days) were released. The T-Plant Canyon was 800 feet long and consisted of a series of cells surrounded by massive concrete walls. T-Plant and two similar plants built during WWII were termed "Queen Mary's" by workers. (Refs. 20, 29, 31 & 128)

December 28, 1944: Connection of B-Reactor's "extra 504" tubes to the cooling water was completed, the reactor loaded and criticality achieved. Plutonium production in quantity had finally begun. (Refs. 31 & 128)

February 1945: F-reactor at Hanford becomes operational, it had a design power level of 250 MWt. It also was graphite moderated; light water cooled reactor with a single-pass cooling system. Three full-scale production reactors (B, D, and F) are now operating. (Refs. 31 & 128)

February 4 – 9, 1945: Yalta conference: Roosevelt, Churchill, and Stalin meet for extensive planning for postwar Europe and Asia; Stalin promises Russian entry into war against Japan two to three months after defeat of Germany. (Refs. 29 & 30)

Early 1945: The S-50 Thermal Diffusion Plant and K-25 Gaseous Diffusion Plant were supplying low-enriched UF₆, which was converted to UCl₄ at Y-12 to be further enriched in the Calutrons. Built in nine months, S-50 fed low-enriched uranium to the Y-12 Plant Calutrons from March 1945 through September 1945. The K-25 gaseous diffusion plant also began feeding LEU to the Y-12 Calutrons in March 1945. Harshaw and Du Pont produced UF₆ from UF₄ as feed for the S-50 Thermal Diffusion and Kr-25 Gaseous Diffusion Plants. (Refs. 19 & 128)

April 1945: B-Plant becomes operational. B-plant was the second chemical separation "canyon" built at the Hanford site for the Manhattan Project. It was used for plutonium recovery from 1945 to 1956. (Refs. 30 & 128)

April 1945: The Soviets enter Berlin. They immediately began to dismantle and ship German industrial equipment to the Soviet Union. (Ref. 32)

April 12, 1945: Roosevelt dies; Truman becomes President. (Refs. 30 & 32)

April 25, 1945: Secretary of War Henry L. Stimson and Groves sneaked into the White House for a 45 minute meeting with President Harry S. Truman to inform him of the Manhattan Project. They sketched the secret research and the timetable for its completion. Truman asked not a single question. (Refs. 30 & 131)

- May 7, 1945: V.E. Day Germany surrenders to Allies ending WWII in Europe. (Refs. 30, 128 & 131)
- May 12, 1945: Franck Report is delivered to Office of Secretary of War, and its substance is communicated to Scientific Advisory Panel. (Ref. 30)
- **June 22, 1945:** Japanese Supreme War Council approves effort to negotiate peace and to seek Russian mediation. (Ref. 30)
- **1945:** The HEU (also called "Oralloy" for <u>Oak Ridge Alloy</u>) from the Calutrons was converted at Y-12 into UF₄ and sent to Los Alamos. The Los Alamos Chemistry and Metallurgy Division further purified the HEU and reduced it to metal for the "Little Boy" atomic bomb. (Refs. 19 & 128)
- **July 1945:** MED acquired part of Oxnard field (now Kirtland Air Force base) in Albuquerque, New Mexico and converted it into weapons assembly site (Sandia Base). (Ref. 128)
- July 12, 1945: Japanese Foreign Minister Togo instructs Ambassador Sato in Moscow to implore Russians to mediate end to war. (Ref. 30)
- **July 14, 1945:** U-235 component for "Little Boy" leaves Los Alamos for transport to Tinian. (Refs. 29 & 32)
- July 16, 1945, 5:30 A.M.: U.S. detonates the first A-bomb (nuclear device was code-named the "Gadget") near Alamogordo, New Mexico. Called "Trinity" test, it explodes with a force equivalent to 21,000 tones of TNT. (Refs. 19, 29, 30, 31, 32, 128 & 131)
- **July 16, 1945:** "Little Boy" —minus the last necessary bit of U-235 was put aboard the cruiser *Indianapolis*, which sailed almost immediately. (Ref. 29)
- July 17 August 2, 1945: Potsdam Conference: First wartime meeting of Truman, Stalin, and Churchill (replaced by Clement Attlee, British prime minister elected during the conference); inconclusive talks about postwar settlements; on July 24, Truman informs Stalin that United States has developed a "new weapon of unusual destructive force." By this time the Soviet Union had a serious atomic bomb project underway. (Refs. 30 & 130)
- July 24, 1945: Sufficient uranium shipped to Los Alamos from Oak Ridge for the manufacture of the first bomb to be dropped on Japan. (Refs. 29 & 32)
- July 25, 1945: US General Carl Spaatz, Commander of the Strategic Air Force in the Pacific, receives directive from Truman ordering 509 Composite Group to "deliver its special bomb as soon as weather will permit visual bombing after about 3 August 1945. (Refs. 30 & 131)
- July 26, 1945: Cruiser Indianapolis arrives in Tinian and discharged "Little Boy." (Ref. 29)
- July 26, 1945: Potsdam declaration is issued. The Potsdam Declaration threatens "prompt and utter destruction" of Japan's hoe islands if an unconditional surrender did not come. (Ref. 30)

July 28, 1945: Japanese Prime Minister Suzuki calls Potsdam Declaration "unworthy of notice." (Ref. 30)

July 30, 1945: Cruiser Indianapolis, in route from Tinian to the Philippines with 900 crew aboard, sunk by Japanese submarine. (Ref. 29)

July 31, 1945: "Little Boy" was ready at Tinian awaiting favorable weather. (Ref. 29)

August 6, 1945: The "Little Boy" A-bomb explodes at 1,900 feet above Hiroshima, with a force equivalent to 12,500 tones of TNT. The bomb was dropped at 0915:30 Tinian Time (August 5, 6:15:30 P. M. Washington time). The "Little Boy" bomb dropped on Hiroshima was a uranium gun-type weapon in which two masses of highly enriched uranium were forced together very quickly to assemble a "critical mass" that would sustain a nuclear chain reaction and subsequent explosion. By the end of the year, bomb-related deaths in Japan totaled 140,000. (Refs. 19, 29, 30, 128, 130 & 131)

August 6 and 7, 1945: The existence of the Manhattan Project and the atomic bomb was revealed to the public after the destruction of Hiroshima when the first public reports on the Manhattan Project issued by the War Department. (Refs. 28 & 128)

August 7, 1945: Stalin put his secret-police chief Laurenti P. Beria in charge of the Soviet version of the "Manhattan Project." (Ref. 130)

August 8, 1945: Russia declares war on Japan. (Ref. 30)

August 1945: Following the bombing of Hiroshima and Nagasaki, Stalin ordered his deputies and Kurchatov to provide "us with atomic weapons in the shortest possible time." (Ref. 130)

August 9, 1945: The "Fat Man" A-bomb explodes at 1,650 feet over Nagasaki with a yield equivalent to 22,000 tons of TNT. The "Fat Man" bomb dropped on Nagasaki was a plutonium implosion bomb that used high explosives to squeeze together a sphere of plutonium very quickly and symmetrically into a "critical mass" that would sustain a nuclear chain reaction and subsequent explosion. 70,000 die in Nagasaki by the end of 1945 from the effects of the bomb. (Refs. 19, 30, 128, 130 & 131)

August 9, 1945: Russia invades Manchuria. (Ref. 30)

August 10, 1945: Japan offers to surrender. (Ref. 30)

August 14, 1945: Japan accepts Allied terms of surrenders. WWII is over. Soon after V-J Day, Du Pont was paid the entire fee of one dollar for its part of the Manhattan Project. Government auditors disallowed this, since the entire time of the contract had not run out. Consequently, Du Pont was asked to return thirty-three cents to the United States. Fortunately, the officers of Du Pont had retained their sense of humor. (Ref. 28 & 30)

-43- 4/15/99

August 1945: The K-25 Plant began producing weapons-grade uranium. The full-scale K-25 gaseous diffusion plant (2,996 diffusion steps or stages) was completed and fully operational at Oak Ridge in August 1945. Large amounts of electricity are required to pump the UF₆ through the diffusion cascade and to remove the heat of compression. (Refs. 19 & 128)

August 30, 1945: U. S. occupation forces land in Japan. (Ref. 30)

September 2, 1945: Formal surrender of Japan. (Ref. 30)

1945: S-50 uranium enrichment plant shuts down. (Ref. 128)

September 20, 1945: US Joint Chiefs of Staff embrace "first-strike" atomic warfare policy. (Ref. 131)

October 1945: United Nations (UN) Created at San Francisco, California. (Refs. 30 & 128)

October 1945: At the Los Alamos scientific laboratory in New Mexico, physicist Edward Teller seeks J. Robert Oppenheimer's (Oppie) support on a full tilt effort to build a hydrogen bomb. Oppenheimer refuses. (Ref. 131)

October 18, 1945: Lavrentii, head of the Soviet secret police and in charge of the Soviet nuclear program, is provided top-secret details on the U. S. plutonium bomb by a spy working inside Los Alamos. (Ref. 131)

November 1945: The original plutonium production site, DP-Site (also known as TA-21) was built. Los Alamos received plutonium nitrate paste from the Hanford site and highly enriched uranium tetrafluoride from the Oak Ridge Y-12 plant. Nuclear components continued to be manufactured at DP-site until the start-up of the Plutonium Finishing Plant at Hanford In July 1949, and the beginning of HEU casting and machining at Y-12 in 1948. (Ref. 128)

November 23, 1945: The USSR concludes a secret agreement with Czechoslovakia granting the Soviet Union exclusive rights to all uranium mined within Czechoslovakia. (Ref. 131)

December 24, 1945: U. S. Embassy in Moscow warns of an all-out effort by USSR to build atomic bombs. (Ref. 131)

January 1946. To meet the projected demand for enriched uranium, AEC expanded the K-25 Gaseous Diffusion Plant in stages beginning in January 1946—another 1,540 stages were added. The increased capacity was completed by June 1954. (Ref. 128)

February 9, 1946: Joseph Stalin gives a speech at the Bolshoi Theater that marks deterioration in relations with the United Station. (Ref. 131)

February 16, 1946: Columbia faculty urges President Truman to stop production of atomic bombs. (Ref. 131)

-44- 4/15/99

February 22, 1946: US Charge d'affaires in Moscow George Kennan sends historic 8,000 word telegram to the State Department. It analyzes Soviet foreign policy in alarming terms. (Ref. 131)

March 5, 1946: Winston Churchill delivers "iron curtain" speech at Fulton, Missouri. (Ref. 131)

April 18, 1946: The secret, three-day Super conference, in New Mexico examines feasibility of developing the hydrogen bomb. (Ref. 131)

April 1946: Soviet scientist Iulii Khariton chooses Sarov/Arzamas as the secret location for Soviet weapons lab. (Ref. 131)

June 14, 1946: British physicist Klaus Fuchs leaves Los Alamos to return to England. (Ref. 131)

July 1946: US conducts atomic test at Bikini Atoll in the South Pacific. This was the initial site of MED and AEC nuclear weapons testing following the end of World War II. (Ref. 128 & 131)

1946: The Salt Wells Pilot Plant at China Lake Naval Ordnance Station in California began production of high explosive main charges. (Ref. 128)

1946: Y-12 uranium enrichment plants shuts down. (Ref. 128)

1946: William L. Borden published a small but influential book on defense strategy in the atomic age – "There Will Be No Time." (Ref. 28)

1946: The Mound Laboratory in Miamisburg, Ohio was built to manufacture polonium-beryllium initiators and other weapons parts. (Ref. 128)

1946: First plutonium-fueled reactor (Clementine) becomes operational at Los Alamos. (Ref. 132)

1946: To limit radiation damage to the reactor's core, the B Reactor at Hanford was shut down. (Ref. 28)

August 1, 1946: President Truman signs Atomic Energy Act that establishes the Atomic Energy Commission (AEC). (Refs. 19, 28, 128 & 131)

September 1, 1946: Du Pont leaves Hanford, General Electric Company becomes the prime site contractor. GE renames the Hanford Site to the "Hanford Atomic Products Operation in 1953 but the name Hanford Works stayed until the coming of the Energy Research and Development Administration in 1975. (Ref. 28)

September 5, 1946: FBI questions US scientist J. Robert Oppenheimer about his contacts with a communist, Professor Haakon Chevalier. (Ref. 131)

-45- 4/15/99

November 10, 1946: Team of Soviet scientists, headed by Igor Kurchatov, begins assembly of first full-scale nuclear reactor in the USSR. (Refs. 130 & 131)

December 1946: B-reactor is closed to preserve it as a future source of polonium-210, the essential initiator material in early atomic explosions. Reactor was shut down to limit the accumulation of radiation-induced swelling and distortion of its graphite core. (Refs. 28 & 128)

December 25, 1946: Soviet scientists achieve nuclear chain reaction. (Refs. 130 & 131)

December 25, 1946: Soviet scientist review espionage accounts of US physicist Edward Teller's "classical super," his design for the hydrogen bomb. (Ref. 131)

Late 1946: General Groves sited the nation's first three national laboratories – Argonne (Illinois), Brookhaven (New York), and Oak Ridge (Tennessee). Funding provided in fiscal 1947 budget. (Ref. 28)

December 31, 1946: Soviet scientist review espionage accounts of US physicist Edward Teller's "classical super," his design for the hydrogen bomb. (Ref. 131)

January 1, 1947: The new AEC takes control of the US atomic weapons complex. The Hanford Engineering Works was renamed the Hanford Works. (Refs. 28, 31 & 128)

March 1947: President Truman "declared" the "Cold War" in a speech calling for military and economic aid to Greece and Turkey, countries he said were struggling against stealthy Soviet intervention. Actually, it can be argued that the "Cold War" began when the Soviet Union began developing its own atomic bomb. (Ref. 28)

March 1947: The Joint Chiefs of Staff of the armed services declared that the weapons supply was "not adequate." (Ref. 131)

1947: The Army Ordnance Plant in Burlington was converted to a nuclear weapons plant. This plant also began manufacture of high explosive main charges. (Ref. 128)

June 5, 1947: US Secretary of State George Marshall announces aid plan for Europe. (Ref. 131)

July 1947: The US passed the National Security act, and Policy Planning Staff Chief George F. Kennan penned his famous article advocating the "containment" of communism and other Soviet influence. (Ref. 131)

August 1947: GE was directed to build two new plutonium production reactors (H and DR) at Hanford and to develop the new reduction oxidation separations process as quickly as possible. (Ref. 28)

September 28, 1947: British physicist Klaus Fuchs meets with his agent Alexander Feklisov in London. Fuchs describes certain structural characteristics of the superbomb. (Ref. 131)

-46- 4/15/99

1947: After engineers discovered a method of reversing the radiation-induced swelling and distortion of the graphite core, AEC authorized the restart of B-Reactor. (Ref. 128)

October 1947: Joint Chiefs of Staff declares that 150 "Nagasaki type" bombs will suffice to defend the US and defeat the USSR. Stockpile of weapons is small (20 to 50) but still growing. (Ref. 131)

December 1947: K-25 began refining it's own UF₆ feed when the F2 Plant became operational. (Ref. 128)

December 3, 1947: The British philosopher and outspoken pacifist, Bertrand Russell, calls for preventive war against USSR. (Ref. 131)

December 1947: Cooling times of irradiated fuel was increased to allowing additional natural decay of radioactive isotopes, especially iodine-131. (Ref. 28)

January 7, 1948: US and Great Britain revoke wartime pact on nuclear cooperation. (Ref. 131)

February 25, 1948: Soviet forces occupy Prague. (Refs. 28 & 131)

March 13, 1948: British physicist Klaus Fuchs gives his agent Alexander Feklisov a detailed description of the "classical super", a design to build the hydrogen bomb. (Ref. 131)

April 1948: AEC begins "Operation Sandstone" at Eniwetok Atoll in the south Pacific to test improved designs of fission bombs. (Ref. 131)

1948: The Kyshtym Complex (also known as Chelyabinsk-40, later called Chekyabinsk-65, it includes the Mayak Chemical Combine), located among the lakes in the upper Techa River area, was constructed in the 1945 – 1948 period. All five plutonium production reactors have been permanently closed. Two tritium production reactors are still operating. (Refs. 25, 26, 27 & 130)

May 5, 1948: President Truman is briefed on the Joint Chiefs of Staff's nuclear war plan. Dubbed "Halfmoon," the plan calls for dropping 50 atomic bombs on 20 Russian cities. Truman disapproves. (Ref. 131)

June 7 (19?), 1948: Reactor A (or Unit 0) at Chelyabinsk-40 (or Kyshtym) reaches full criticality, enabling the USSR to produce plutonium. Unit 0 was a graphite-moderated, natural-uranium-fueled (aluminum-clad) reactor; it was loaded with all the uranium then available in the country. Construction of A-Reactor, called "Anotchka," or "Little Anna" in English, began around 1946. Shutdown occurred in 1987, after 39 years of operation. The initial power level was 100 MWth and the final power level was 500 MWth. Cooling water from a nearby lake was pumped directly through the core. (Refs. 25, 26, 27, 130 & 131)

June 1948: Soviet physicist Igor Tamm enlists his graduate student Andrei Sakharov to study fusion problem. (Ref. 131)

-47- 4/15/99

June 24, 1948: USSR blocks rail and road connections to West Berlin (The Berlin Blockade). The US responded with the Berlin airlift (an 11-month ordeal). (Refs. 28 & 131)

July 1948: Soviet physicist Andrei Sakharov begins development of "Layer Cake" concept for hydrogen bomb. (Ref. 131)

1948: AEC institutes an incentive program to stimulate the domestic mining and milling of uranium. (Ref. 128)

1948: Enewetak Atoll (formerly spelled Eniwetok) in the South Pacific opened for nuclear testing. (Refs. 28 & 128)

1948: The Y-12 Plant at Oak Ridge, Tennessee began making uranium weapons parts. (Ref. 128)

1948: Heavy-water production commences at the Chirchik nitrogen plant. Here the method was to obtain heavy water as a by-product from the synthetic ammonia industry. (Refs. 25, 26 & 27)

1948: B Reactor at Hanford restarted. (Ref. 28)

October 19, 1948: General Curtis LeMay assumes command of the Strategic Air Command. (Ref. 131)

January 1949: Soviet physicist Andrei Sakharov moves to Arzamas, the secret weapons laboratory. (Ref. 131)

February 1949: Secret negotiations between the USSR and Western Allies begin to solve the Berlin crisis. (Ref. 131)

February 27, 1949: Plutonium separations begin at Chelyabinsk. (Ref. 131)

March 1949: The first General Curtis LeMay war plan for the Strategic Air Command envisions attacks on 70 Soviet cities with 133 bombs. (Ref. 131)

May 1949: Chinese Communists take power in China. (Refs. 128 & 131)

May 15, 1949: Communists win election in Hungary. (Ref. 131)

July 1949: Hanford takes over from Los Alamos the manufacturing of plutonium pits at the Plutonium Finishing Plant (PFP). (Ref. 128)

1949: Hanford Works planners initiated completion of "C" Plant in 200 East Area as a "hot semi-works" (pilot plant) for the REDOX process. C Plant had been started in 1944 as the fourth separations canyon but not finished. (Ref. 128)

1949: Hanford begin manufacturing lithium targets for tritium production. (Ref. 128)

-48- 4/15/99

1949: The Soviet's first gaseous diffusion cascade was constructed in Kefirstadt (now called Verkhniy-Neyvinskiy). (Ref. 128)

1949: Assembly functions performed at Sandia Base were transferred to the Burlington assemble plant. (Ref. 128)

1949: The Kansas City Plant in Missouri began making nonnuclear weapons parts (electronics, rubber, plastic, foams, adhesives, outer casings, and others). (Ref. 128)

1949: Electromet in Niagra Falls, NY, discontinued production of UF₄ and uranium metal. (Ref. 128)

1949: Both the North Atlantic Treaty Organization (the US and its allies) and the Warsaw Pact (USSR and its satellites) are formed. (Ref. 28)

August 29, 1949: First Soviet atomic bomb is exploded in Kazakhstan (Joe 1) at the Semipalatinsk test range. Plutonium from Unit 0 at Kyshtym was used for this bomb. (Refs. 28, 128, 129, 130, 131 & 132))

September 3, 1949: A US weather plane flying off the coast of Siberia picks up evidence of radioactivity. (Ref. 131)

September 23, 1949: President Truman announces explosion of first Soviet atomic bomb. (Ref. 131)

October 1949: US government approves expanded production of uranium and plutonium. (Ref. 131)

October 1949: Operations at "H" reactor begin at Hanford. It was a graphite-moderated, lightwater cooled reactor with single-pass cooling system. H-Reactor was designed to operate at 400 MWt. (Refs. 28 & 31)

October 29, 1948: General Advisory Committee to the AEC argues against a crash program to develop the hydrogen bomb. (Ref. 131)

November 1, 1949: Sandia Laboratory was formed from the Sandia Branch of Los Alamos on the grounds of Oxnard Field (now Kirtland Air Force base) near Albuquerque, New Mexico. The mission of the new laboratory was the design of nonnuclear components of weapons. (Ref. 128)

Early November 1949: Kurchatov begins work on the development of a thermonuclear bomb as a matter of priority. (Ref. 131)

November 25, 1949: AEC Commissioner Lewis Strauss writes to President Truman urging him to give highest priority to hydrogen bomb development. (Ref. 131)

January 1950: Alger Hiss convicted of spying. (Ref. 28)

January 27, 1950: In London British physicist Klaus Fuchs confesses to being a Soviet spy. (Refs. 28 & 131)

January 31, 1950: President Truman announces decision to develop Thermonuclear Weapons — the hydrogen bomb. (Refs. 28, 128 & 131)

February 5, 1950: Twelve leading US physicists speak out against President Truman's decision to build the hydrogen bomb. (Ref. 131)

February 1950: Wisconsin Senator Joseph McCarthy launched four years of accusations that communists infested the American government and media. (Ref. 131)

February 1950: Joint Intelligence Committee predicts build up of Soviet atomic arsenal and possible attack against US "at earliest possible moment." (Ref. 131)

February 24, 1950: US Joint Chiefs of Staff request "all out effort to build H-Bomb." (Ref. 131)

March 1, 1950: British physicist Klaus Fuchs is tried at the Old Bailey in London for being a soviet spy. (Ref. 131)

March 1950: Mao Tse-tung's Communist forces drive Nationalist forces under Chiang Kaishek from Mainland China. (Ref. 28)

April 7, 1950: National Security Council document NSC-68 warns of surprise attack by Soviet Union once "it has sufficient atomic capability." (Ref. 131)

April 1950: Mao Tse-tung signed a mutual assistance pact with Soviet dictator Josef Stalin. (Ref. 28)

June 16, 1950: US mathematicians Stanislaw Ulam and Cornelius Everett conclude their calculations on "classical super," Edward Teller's H-Bomb design. The plan appears not to work. (Ref. 131)

June 25, 1950: Communist North Korean forces crossed south of the 38th parallel, invading South Korea and igniting the Korean Conflict. The US, quickly assuming a lead in international affairs, supplied 90 percent of the troops fighting North Korea. (Refs. 28 & 131)

1950: The Plutonium Finishing Plant (PFP, 234-5 Z building) at Hanford began converting plutonium nitrate into more stable plutonium oxide and metal. (Ref. 128)

1950: Y-12 Plant at Oak Ridge was tasked with the development of lithium isotope separation technically. (Ref. 128)

September 30, 1950: The NSC-68 document that warns of surprise attack by the Soviet Union is adopted as statement of policy. Defense spending is increased by more than 350%. (Ref. 131)

-50- 4/15/99

October 1950: Operations at "DR" reactor begin. It was a graphite-moderated light-water cooled reactor with a single pass cooling system. DR-reactor was designed to operate at 250 MWt. (Refs. 28 & 31))

November 30, 1950: President Truman confirms during press conference that use of nuclear weapons in Korea had been under consideration. (Ref. 131)

December 16, 1950: With the Chinese army having entered the Korean war, President Truman declares national emergency and signs order to increase armed forces by 3.5 million men. (Ref. 131)

Late 1950 or early 1951: Additional heavy water production plants go into operation at Kirovakan, Dneprodzerzhinsk, and Gorlovka. These plants also recovered heavy water as a byproduct of synthetic ammonia production. (Ref. 25, 26, 27 & 130)

January 1951: US mathematician Stanislaw Ulam proposes radical new design for H-Bomb. Edward teller embraces and refines the concept. (Ref. 131)

January 12, 1951: President Truman establishes the Federal Civil Defense Administration. (Ref. 131)

April 4, 1951: US physicist Edward teller submits report on new design for H-Bomb. (Ref. 131)

1951: AEC established the Feed Materials Production Center in Fernald, Ohio. (Ref. 128)

April 5, 1951: US Joint Chiefs of Staff order atomic retaliation against air bases in case of a "a major attack" against UN forces in Korea. (Ref. 131)

May 9, 1951: US conducts the "George" test in the Pacific: a large fission bomb that triggers the first thermonuclear reaction. (Ref. 131)

1951: The Pantex Plant, near Amarillo, Texas was converted from a conventional munitions plant to a nuclear weapons assembly plant. This plant also manufactured high explosive main charges. (Ref. 128)

1951: The REDOX Plant at Hanford (also known as S-Plant) began operations. (Ref. 128)

1951: Harshaw discontinued production of UF₄. (Ref. 128)

1951: The Rocky Flats Plant begins operations. (Refs. 28 & 128)

1951: Nevada Test Site opens. This 1,350-square-mile area of the southern Nevada desert has been the site of most of the US underground and atmospheric testing. There have been 928 nuclear tests, including 100 atmosphere tests. (Ref. 128)

1951: AEC establishes the Savannah River Site near Aiken, South Carolina. (Refs. 28 & 128)

-51- 4/15/99

September 24, 1951: Soviet Union conducts its second nuclear test, an improved plutonium bomb; yield was at least 25 kilotons. (Ref. 131)

October 18, 1951: The Soviet's test their first nuclear device utilizing highly enriched uranium; yield about 50 kilotons. (Ref. 131)

December 1951: A four-man team at RAND begins to study the likely effects of the H-Bomb. (Ref. 131)

1952: Three reactors are operational at Chelyabinsk-40.

January 1952: The reduction oxidation (REDOX) processing plant, the first continuous solvent extraction plant, was completed and began operations. REDOX used methyl isobutyl ketone (known as hexone) as the organic extractant and aluminum nitrate as the "salting agent." (Refs. 28 & 128)

February 1952: Livermore, California was selected as the site for a second, dual capability, nuclear weapons design and R&D laboratory. (Ref. 128)

March 1952: The Royal Air Force and the Strategic Air Command begin flying photographic and radar reconnaissance missions over Soviet Union. (Ref. 131)

Summer 1952: The initial operations of REDOX brought the first self-boiling wastes to Hanford's waste tanks. S tank Farm contents began to boil in the summer of 1952, but the real problems arose the following winter, as cooling mechanisms not designed for self-boiling wastes froze and broke. A "swamp" of contaminated condensate and flooded cooling water, along with loud "bumps" from thermal releases in the tanks caused problems throughout the winter of 1952-1953. (Ref. 28)

1952: The Rocky Flats Plant near Golden, Colorado began manufacturing plutonium, high enriched uranium and depleted uranium pit parts. Rocky Flats assembled parts from Hanford, Y-12, and the South Albuquerque Works into completed pits. (Refs. 28 & 128)

1952: The OREX lithium isotope enrichment pilot plant constructed at Y-12, Oak Ridge. (Ref. 128)

1952: Feed Materials Production Center (FMPC) at Fernald, Ohio opens and begins refining uranium. Uranium refining is consolidated at the Mallinckrodt Chemical Works in St. Louis and FMPC. (Ref. 28 & 128)

1952: B Plant (bismuth phosphate process) at Hanford shut down. (Ref. 28 & 128)

1952: Hanford stops manufacturing lithium targets for tritium production. (Ref. 128)

1952: Steel Component fabrication functions were moved from across the nation to the South Albuquerque Works in New Mexico. (Ref. 128)

-52- 4/15/99

- 1952: The heavy water plants at the Savannah River Site, South Carolina, and the Dana Plant at Newport, Indiana began operations to supply large amounts of heavy water for the Savannah river Site reactors. The Dana Plant operated from April 1952 until May 1957; the Savannah River Heavy Water Plant began operations in October 1952. (Ref. 128)
- 1952: M-Area at Savannah River built to clad and assemble fuel elements for the five production reactors located there. (Ref. 128)
- 1952: Construction of the Portsmouth Gaseous Diffusion Plant at Piketon, Ohio begins. (Refs. 28 & 128)
- **September 1952:** AEC officially opened the University of California Radiation Laboratory in Livermore, California as a second nuclear design laboratory. The facility is now known as the Lawrence Livermore National Laboratory. (Refs. 128, 131 & 132)
- October 3, 1952: First British nuclear test, code-named "Hurricane," is conducted off the northwest coast of Australia. (Ref. 131)
- **November 1, 1952:** "Mike," the first US Experimental Thermonuclear Device i.e., the first H-bomb experiment, is successfully tested at Eniwetok in the Pacific (10.4 megatonnes). (Refs. 128 & 131)
- **November 1952:** C Reactor at Hanford goes critical, it had a design power level of 600 MWt. It, also, was a graphite-moderated light-water cooled reactor with a single pass cooling system. (Refs. 28 & 31)
- 1952: U Plant, built at Hanford during WWII but not needed as a processing canyon, was retrofitted as the Metal Recovery Plant. Its mission was to use a tri-butyl phosphate in saturated kerosene (TBP-NPH) extraction technique to recover uranium from the waste stored in the tank farm. At the time, the scarcity of high-grade uranium supplies made this mission crucial. Much of the US supply of uranium was housed in Hanford's waste tanks. (Ref. 28)
- **December 1952:** President-elect Eisenhower and staff develop "New Look" defense policy relying primarily on power of atomic forces. Eisenhower believed that the US needed a huge, single strike capacity. This policy was termed "massive retaliation." (Refs. 28 & 131)
- **January 1953:** In his final State of the Union address, President Truman declares nuclear impossible for "rational men." (Ref. 131)
- **January 1953:** Construction of the 1,812 enrichment stages of the Paducah Plant, in Paducah, Kentucky, begins. (Ref. 28)
- 1953: Production-scale lithium enrichment using the ELEX Process began operation at the Y-12 Plant, Oak Ridge, Tennessee. (Ref. 128)
- 1953: The first Soviet production-model tactical atomic bomb tested at the Semipalatinsk test range. This weapon was in service from 1954 to 1965. (Ref. 129)

-53- 4/15/99

March 5:1953: Soviet leader Joseph Stalin dies. (Refs. 130 & 131)

March 20, 1953: Nikita Khrushchev becomes first secretary of Communist party. (Refs. 28, 130 & 131)

July 27, 1953: Armistice is signed ending war in Korea. (Ref. 131)

1953: The Idaho chemical Processing Plant (ICPP) at the Idaho National Engineering Laboratory began using variants of PUREX to process spent navy and experimental reactor fuel for recovery and recycle of high enriched uranium (HEU). (Refs. 28 & 128)

August 1953: General Edmundson leads "Operation Big Stick." The mission required him to take B-36s, armed with nuclear weapons, to Okinawa in Japan. (Ref. 131)

August 8, 1953: Soviet Premier Georgii Malenkov announces that USSR possesses hydrogen bomb. (Ref. 131)

August 12, 1953: First Soviet Thermonuclear Device ("Joe 4") is tested. This device was Andrei Sakharov's "Layer Cake" device that utilized lithium deuteride as the fusion material. (Refs. 28, 128, 130 & 131)

December 1953: R-reactor at Savannah River goes critical. This production reactor used heavy water as a moderator and primary cooling medium. The primary coolant was completely contained in the reactor building. Heat was extracted through the use of heat exchangers cooled by water from the Savannah River. R-Reactor was designed to operate at less than 500 MWt but during the period from 1955 to 1965, the thermal power level was increased to approximately 2500 MWt by engineering enhancements. (Refs. 31 & 128)

December 1953: President Eisenhower announces the "Atoms for Peace" Program. (Refs. 28, 31 & 128)

January 12, 1954: Secretary of State John Foster Dulles announces administration policy of "massive retaliation" in response to Communist attacks. (Ref. 131)

1954: Plutonium and tritium production reactors startup at the Savannah River Site. (Ref. 128)

1954: Mound began tritium work. (Refs. 128)

1954: The Atomic Energy Act (AEA) was amended to encourage the peaceful use of atomic energy. (Refs. 31 & 128)

1954: M-area at Savannah River began manufacturing fuel for the Savannah River Rachers. (Ref. 128)

February 1954: Soviet physicists Andrei Sakharov and Igor Tamm are presented with the Hero of Socialist Labor and the Stalin Prize for their work on the "Layer Cake." (Ref. 131)

-54- 4/15/99

February 1954: P-reactor at Savannah River goes critical. This production reactor used heavy water as a moderator and primary cooling medium. The primary coolant was completely contained in the reactor building. Heat was extracted through the use of heat exchangers cooled by water from the Savannah River. P-Reactor was designed to operate at less than 500 MWt but during the period from 1955 to 1965, the thermal power level was increased to approximately 2500 MWt by engineering enhancements. (Refs. 31 & 128)

February 1954: "Castle Bravo" test—The largest US test at 15 megatonnes. (Refs. 128 & 131)

July 1954: L-reactor at Savannah River goes critical. This production reactor used heavy water as a moderator and primary cooling medium. The primary coolant was completely contained in the reactor building. Heat was extracted through the use of heat exchangers cooled by water from the Savannah River. L-Reactor was designed to operate at less than 500 MWt but during the period from 1955 to 1965, the thermal power level was increased to approximately 2500 MWt by engineering enhancements. (Refs. 31 & 128)

Fall 1954: The Salt Wells Pilot Plant at China Lake Naval Ordnance Station in California stopped production of high explosive main charges. (Ref. 128)

September 14, 1954: 44,000 Soviet troops take part in a military exercise involving the dropping of an atomic bomb. (Ref. 131)

October 1954: K-reactor at Savannah River goes critical. This production reactor used heavy water as a moderator and primary cooling medium. The primary coolant was completely contained in the reactor building. Heat was extracted through the use of heat exchangers cooled by water from the Savannah River. K-Reactor was designed to operate at less than 500 MWt but during the period from 1955 to 1965, the thermal power level was increased to approximately 2500 MWt by engineering enhancements. (Refs. 31 & 128)

November 1954: F-Canyon at the Savannah River Plant becomes operational. This was the first large-scale use of the PUREX Solvent Extraction Process for the separation and purification of plutonium and uranium. (Ref. 128)

December 1954: Construction of the 1,812 enrichment stages of the Paducah Plant, in Paducah, Kentucky, is completed. (Ref. 128)

January 1955: KW Reactor at Hanford goes critical, it had a design power level of 1800 MWt. It, also, was a graphite moderated, light water cooled reactor with single-pass cooling system. (Refs. 28, 31 & 128)

March 1955: C-reactor at Savannah River goes critical. This production reactor used heavy water as a moderator and primary cooling medium. The primary coolant was completely contained in the reactor building. Heat was extracted through the use of heat exchangers cooled by water from the Savannah River. C-Reactor was designed to operate at less than 500 MWt but during the period from 1955 to 1965, the thermal power level was increased to approximately 2500 MWt by engineering enhancements. (Refs. 31 & 128)

April 1955: KE Reactor at Hanford goes critical, it had a design power level of 1800 MWt. It, also, was a graphite moderated, light water cooled reactor with single-pass cooling system. The coming to power of "KE" and "KW" reactors brought the total number of reactors operating at Hanford to eight. KE and KW were called the "jumbo" reactors, because their nameplate design power levels was 1,800 megawatts, whereas the earliest Hanford reactors were at about one-fifth that level. (Refs. 28, 31 & 128)

May 10, 1955: The Soviet Union unexpectedly accepts UN proposal for nuclear disarmament. (Ref. 131)

May 14, 1955: Warsaw Pact is signed. (Ref. 131)

1955: The Recuplex operation, a method of reclaiming plutonium from various process scraps and waste, was started up at Hanford. (Refs. 28 & 128)

1955: The first Savannah River tritium facility was built in F-Area to recover tritium from irradiated lithium-6 targets. This facility also began loading tritium into weapons components. (Ref. 128)

1955: Two large COLEX production-scale lithium enrichment plants were built. Alpha 4 began operations in January 1955; Alpha 5 began operations later that same year. (Ref. 128)

June 15, 1955: President Eisenhower evacuates the White House in Operation Alert air raid drill. (Ref. 131)

July 1955: H-canyon at Savannah River begins operation. (Ref. 128)

July 18, 1955: Big Four summit begins in Geneva. President Eisenhower unveils his proposal for "open skies" and an exchange of military secrets. (Ref. 131)

August 8, 1955: In Geneva, the first UN conference begins on the peaceful use of atomic energy. (Ref. 131)

Late 1955: The PUREX Plant at Hanford began test runs. (Ref. 128)

September 6, 1955: US delegate Harold Stassen announced that America no longer supports UN plan calling for complete nuclear disarmament. (Ref. 131)

November 22, 1955: The first Soviet two-stage thermonuclear device tested. Bomb is dropped from an aircraft in Kazakhstan. (Ref. 131)

January 1956: The PUREX Plant at Hanford began hot operations. (Refs. 19, 28 & 128)

February 1956: Construction of the 4,080 stages of the Portsmouth Gaseous diffusion Plant is completed. (Refs. 28 & 128)

February 14, 1956: Nikita Khrushchev attacks Stalin and "cult of personality." (Ref. 131)

-56- 4/15/99

March 1956: US explains its opposition to nuclear disarmament at UN stating that atomic weapons are a "powerful deterrent to war." (Ref. 131)

1956: A branch of Sandia National Laboratory was established at Livermore, California. (Ref. 128)

1956: The Weldon Springs plant near St. Louis was converted from a conventional ordnance production facility and began uranium-refining operations. (Ref. 128)

1956: T Plant (bismuth phosphate process) shuts down. (Refs. 28 & 128)

1956: The first significant, confirmed single-shell waste tank leak at Hanford occurred at 104-U (55,000 gallons). (Refs. 28 & 128)

1956: AEC directed the shut-down of CP-2 and CP-3. (Ref. 128)

1956: The ELEX lithium-enrichment plant was shut down. (Ref. 128)

1957: The Dana heavy water plant in Newport, Indiana was shut down. (Ref. 128)

1957: Mallinckrodt discontinued the production of UF₄. (Ref. 128)

1957: The first large-scale nuclear power plant in the world was built at Shippingport, Pennsylvania. (Ref. 19)

1957: Technical Area 2 at Sandia base stops assembly of nuclear weapons. (Ref. 128)

1957: First reactor at Krasnoyarsk-26 was built. The three reactors at Krasnoyarsk-26 are located 200 to 250 meters underground. More than 65,000 prisoners and 100,000 soldiers were required to dig the underground areas. The cooling water for these reactors is taken from and discharged to the Yenisey River. (Ref. 25, 26, 27 & 130)

1957: The Pinellas plant was built in Largo, Florida, to produce precisely timed, accelerator-type neutron generators to initiate chain reactions in nuclear weapons. (Ref. 128)

1957: Mound stopped production of initiators for nuclear weapons. Mound assigned new production functions including detonators, cable assemblies, and firing sets. (Ref. 128)

August 1957: First US underground Nuclear Test. (Refs. 128 & 131)

September 1957: Kyshtym Disaster. Explosion of a high-level liquid waste tank at Chelyabinsk-40 expelling about 2 million curies onto the atmosphere and surrounding environment. This was probably the result of a reaction between nitric acid and acetic acid from the acetate precipitation process used by the Russians to process plutonium targets. The accident resulted in the evacuation and/or resettlement of thousands of people in the area, and in the isolation of Lake Kyzyltash through the construction of a canal and reservoir system. (Refs. 25, 26, 27 & 130)

October 4, 1957: Sputnik I, the world's first man-made vehicle to orbit the earth, was launched by the USSR. Within seven months, the USSR launched Sputniks II and III. (Refs. 28 & 128)

October 1957: International Atomic Energy Agency (IAEA) established. (Refs. 31, 128 & 131)

January 1958: EURATOM established. (Refs. 31, 128 & 131)

1958: The U-Plant at Hanford shuts down. U-Plant was used to recover uranium that had been placed into the high level waste tanks during and after WWII (via the bismuth phosphate process). (Refs. 28 & 128)

1958: The downtown St. Louis uranium refining plant shuts down. (Ref. 128)

1958: US-UK Mutual Defense Agreement. (Ref. 31)

1958: Khrushchev exacerbated world tensions by renewing demands that the North Atlantic Treaty Organization (NATO) abandon their sectors of Berlin. (Ref. 28)

1958: Bikini Atoll closed as a nuclear testing site. Between 1946 and 1958, 23 tests took place at Bikini. (Ref. 128)

1958: Enewetak Atoll closed as a nuclear testing site. Between 1948 and 1958, Enewetak Atoll was used for 43 atmospheric tests, including the first thermonuclear test in 1952. (Ref. 128)

1958: Production of beryllium components becomes part of normal operation at Rocky Flats. (Ref. 128)

1958: Johnston and Christmas Islands opened as atmospheric nuclear weapons testing sites. (Ref. 128)

1958: The second Savannah River tritium facility was built in H-Area to recover tritium from irradiated lithium-6 targets. (Ref. 128)

September 1958: The first Siberian reactor goes on-line. This plant is near Tomsk. In 1976, then chairman of the State committee for the Utilization of atomic Energy, Dollezhal', wrote: "The Siberian nuclear power station is a classic example of the use of waste heat in the production of plutonium for the generation of electric power. The principal expenses of this nuclear power station are covered by the cost of the plutonium production." This light-water cooled, graphite-moderated, power-and-plutonium production, channel-type reactors are designed for on-line refueling. The three least safe of five plutonium production reactors have been shut down. The two operating 2500 MWt light water-cooled, graphite-moderated production reactors (ADE-4 & ADE-5) are to be closed by the year 2000. It is estimated that these reactors produced approximately 70 tonnes of weapons-grade plutonium. (Refs. 25, 26 & 27)

-58- 4/15/99

October 1958: President Eisenhower announced a unilateral US moratorium on nuclear weapons tests with the understanding that the Soviet Union would likewise refrain from conducting nuclear tests. (Refs. 31, 128 & 131)

1959: One of the COLEX lithium-enrichment plants was shut down. (Ref. 128)

1959: During an American visit, Nikita Khrushchev told Americans: "Your grandchildren will live under communism!" (Ref. 28)

December 1959: The second channel-type reactor reaches full power at Tomsk. (Refs. 3, 25, 26, 27 & 29)

1960: The "hot semi-works" (C Plant) at Hanford Site began pilot runs to extract large amounts of strontium (Sr-90) from plant waste. (Ref. 128)

December 1960: The third channel-type reactor reaches full power at Tomsk.

February 1961: First French Nuclear Test. (Refs. 128 & 131)

1961: Khrushchev angrily erects the Berlin Wall. President John f. Kennedy traveled to Berlin to dramatically reaffirm the American resolve to stay in that city, "Ich bin ein Berliner" (I am a Berliner), he proclaimed. (Ref. 28)

1961: Second reactor at Krasnoyarsk-26 was built. The three reactors at Krasnoyarsk-26 are located 200 to 250 meters underground. More than 65,000 prisoners and 100,000 soldiers were required to dig the underground areas. The cooling water for these reactors is taken from and discharged to the Yenisey River. (Refs. 25, 26 & 27)

September 1961: The Soviet Union breaks the nuclear weapons testing moratorium with a series of the largest tests ever conducted. (Refs. 31 & 128)

September 30, 1961: With the nuclear weapons testing moratorium broken, the US resumed nuclear testing at the Nevada Test Site. (Refs. 31 & 131)

October 11, 1961: First Soviet underground nuclear test. (Ref. 130)

November 30, 1961: USSR explodes a 50 megatonnes H-bomb (largest in the world) at the Novaya Zemlya test range. (Refs. 128 & 129)

December 1961: The fourth channel-type reactor reaches full power at Tomsk. (Refs. 3, 25, 26, 27 & 30)

April 7, 1962: An accidental nuclear excursion occurred in the plutonium waste recovery facility (Recuplex) of BLDG 234-5. This criticality was the first and only one to occur in any production facility at Hanford. Three employees received overexposures to gamma and neutron radiation of 110, 43, and 19 rem. This criticality closed the Recuplex operation. Recuplex was replaced by the Plutonium Reclamation Facility. (Ref. 28)

-59- 4/15/99

1962: The UF₆ production plants at K-25, Portsmouth, and Paducah closed down. After that time, commercial suppliers in Metrolpolis, Illinois converted uranium to UF₆ feed. (Ref. 128)

1962: Rocky Flats ceases making HEU components. This leaves Y-12 as the sole source for these components. (Ref. 128)

March 1962: US government ends purchases of uranium ore for weapons programs. In total, MED and AEC purchased over 3.6 million tons of domestic uranium ore, equivalent to 11,373 tons of U3O8 concentrate. AEC domestic and foreign concentrate purchases continued until 1971 and totaled 325,000 tons, consisting of 175,000 tons from domestic sources and 150,000 tons from foreign sources. (Ref. 128)

1962: Johnston and Christmas Islands closed as atmospheric nuclear testing sites. Johnston Island was the site of 12 tests, Christmas Island was the site of 24 tests. (Ref. 128)

October 1962: Cuban "missile crisis." President John F. Kennedy successfully challenged the Soviet attempt to emplace Inter Continental Ballistic Missiles in the Western Hemisphere. (Refs. 28 & 128)

December 1962: The fifth channel-type reactor reaches full power at Tomsk. (Ref. 3, 25, 16, 27 & 130)

1963: The remaining COLEX lithium-enrichment plant was shut down. (Ref. 128)

1963: The United States stopped atmospheric testing of nuclear weapons. (Refs. 19 & 128)

September 1963: President Kennedy spoke at the opening ceremony for the Hanford N Reactor. (Ref. 128)

1963: X-10, the Oak Ridge Graphite Reactor, was decommissioned. It is now a national historic landmark. (Ref. 128)

October 1963: US and USSR sign a limited Test Ban Treaty. (Refs. 128 & 131)

December 1963: N Reactor began weapons production operations at Hanford. This was the last production reactor built at the Hanford site. N reactor began weapons plutonium production operation in December 1963 and started the steam generation of commercial electric power in 1966. N-Reactor had a recirculating primary coolant system and operated at higher pressures and coolant temperatures than the other eight reactors at Hanford. N-Reactor was designed to operate at 4000 MWt. (Refs. 28, 31 & 128)

December 1963: The sixth channel-type reactor reaches full power at Tomsk. (Refs. 3, 25, 26, 27 & 130)

1964: The Plutonium Reclamation Facility opens in Z-Plant at Hanford. (Ref. 128)

April 1964: The first graphite-moderated, channel reactor begins operation at Beloyarskiy. (Refs. 25, 26 & 27)

1964: R-Reactor at the Savannah River Site shuts down permanently. (Ref. 128)

1964: AEC discontinues HEU production for weapons because it had accumulated sufficient stocks. The AEC continued to produce HEU for other AEC programs, including civilian nuclear power research and the US Navy nuclear power program. (Ref. 128)

1964: Third reactor at Krasnoyarsk-26 was built. The three reactors at Krasnoyarsk-26 are located 200 to 250 meters underground. More than 65,000 prisoners and 100,000 soldiers were required to dig the underground areas. The cooling water for these reactors is taken from and discharged to the Yenisey River. (Refs. 3, 25, 26, 27 & 130)

August 26, 1964: Private ownership of Special Nuclear Materials Act. (Ref. 31)

December 1964: H-Reactor at Hanford is shutdown. (Ref. 128)

October 1964: First Chinese Nuclear Test. (Refs. 128 & 131)

April 1965: H-Reactor at Hanford is shutdown. (Ref. 128)

June 1965: F -Reactor at Hanford is shutdown. (Ref. 128)

1965: AEC ends Hanford's plutonium component manufacturing. This leaves Rocky Flats as the sole source of plutonium components. (Refs. 28 & 128)

1966: Hanford's N reactor started the Hanford Generating Project, producing over 65 billion kilowatts of electricity over 24 years. (Refs. 28, 31 & 128)

1966: The South Albuquerque Works closed, transferring its stainless steel pit component and tritium reservoir fabrication mission to rocky Flats. (Ref. 128)

1966: Weldon Springs uranium refining plant closed down. (Ref. 128)

1966: The Nuclear Fuel Services Plant at West Valley, New York, began processing irradiated, slightly enriched uranium fuel. This was the only plant that has processed privately owned nuclear power plant fuel in the United States. It shut down in 1972. (Ref. 128 & 131)

June 1967: D-Reactor at Hanford is shutdown. (Refs. 28 & 128)

June 1967: The REDOX Plant at Hanford shuts down. It processed more than 19,000 tonnes of spent fuel in its lifetime. (Refs. 28 & 128)

1967: Molten salt extraction replaced an anion exchange process for removing 241-Am ingrowth from recycle plutonium. (Ref. 128)

-61- 4/15/99

September 1967: The second graphite-moderated, channel reactor begins operation at Beloyarskiy. (Refs. 25, 26, 27 & 130)

December 1967: The third graphite-moderated, channel reactor begins operation at Beloyarskiy. (Refs. 25, 26 & 27)

1968: The Savannah River Site converted to HEU fuel and DU targets. F-Canyon given mission of processing irradiated Du targets and recoveying ²³⁹Pu as well as Am, Cm, and other isotopes; H-Canyon assigned to process HEU spent fuel and to recover ²³⁵U, ²³⁷Np, ²³⁸Pu. (Ref. 128)

1968: The Hanford B plant was modified and used to remove, encapsulate, and store radioactive cesium (Cs-137) and strontium (Sr-90) from the Hanford high level waste tanks. The strontium and cesium recovery programs were an attempt to separate high-level waste components so that residual liquids could be disposed to the ground. (Refs. 28 & 128)

February 1968: L-Reactor at Savannah River placed in standby. (Ref. 128)

July 1968: Nuclear Non-Proliferation Treaty. (Refs. 128 & 131)

April 1969: C-Reactor at Hanford is shutdown. (Ref. 128)

1969: Mound begins retrieving tritium from retired weapons to be recycled and sent to Savannah River Site for purification and reuse. (Ref. 128)

December 1969: The third graphite-moderated, channel reactor begins operation at Belovarskiy. (Refs. 25, 26 & 27)

1970: Clean Air Act. (Ref. 128)

1970: The Nuclear Nonproliferation Treaty (NPI) enters into force. (Ref. 31)

January 1971: KE-Reactor at Hanford is shutdown. (Refs. 28 & 128)

1971: US government ends purchases of uranium concentrate from both domestic and foreign sources. (Ref. 128)

1971: The name of the University of California Radiation Laboratory—Livermore was changed to Lawrence Livermore Laboratory. (Ref. 128)

May 1972: SALT I Treaty signed. (Refs. 128 & 131)

1972: Clean Water Act. (Ref. 128)

1972: PUREX Plant at Hanford placed into "cold standby" because of excess of separated fuel-grade plutonium. (Refs. 19, 28 & 128)

-62- 4/15/99

1972: The Fernald uranium refinery closed. (Ref. 128)

1973: The largest known leak from a Hanford single-shell tank occurred when 115,000 gallons escaped from 106-T. (Refs. 28 & 128)

May 1974: India detonated a nuclear device, having obtained the necessary plutonium from its own reprocessing plant. (Ref. 131)

December 31, 1974: AEC was abolished by the Energy Reorganization Act. (Ref. 28 & 128)

January 1, 1975: Energy Research and Development Administration (ERDA) takes over the "Nuclear Weapons Complex" from the AEC. The Nuclear Regulatory Commission (NRC) takes charge of regulating the civilian uses of atomic energy. (Refs. 19, 28 & 128)

1975: The Burlington Assembly Plant shuts down and transfers its functions (weapon assembly and manufacture of high explosive main charges) to the Pantex Plant. (Ref. 128)

1976: "B" reactor at Hanford was named as a National Mechanical Engineering Landmark by the American Society of Mechanical Engineers. (Refs. 28 & 128)

October 1976: Resource Conservation & Recovery Act (RCRA). (Ref. 128)

October 28, 1976: President Ford declared, "the avoidance of proliferation must take precedence over economic interests" and declared that US policy must be changed to deferral "of the commercialization of chemical reprocessing of nuclear fuel..." (Ref. 131)

April 1977: President Carter deferred indefinitely the commercial reprocessing and recycling of plutonium produced in the US nuclear power programs. (Ref. 131)

1977: The newly created Department of Energy assumes the duties of the ERDA. (Refs. 19 & 128)

1978: TA-55 at Los Alamos replaces the original plutonium production site, DP-Site. (Ref. 128)

1978: Nuclear Non-Proliferation Act in the US Congress

November 1978: Uranium Mill Tailing Radiation Control Act. (Ref. 128)

March 1979: Three Mile Island Accident. (Ref. 128)

1980: The PFP at Hanford shut down. (Refs. 28 & 128)

December 1980: Comprehensive Environmental Response Compensation & Liability Act (CERCLA). (Ref. 128)

1981: C-Reactor at Savannah River dedicated to tritium production. (Ref. 128)

-63- 4/15/99

1981: President Reagan lifted the indefinite ban on commercial reprocessing activities in the US. (Ref. 131)

1981: At the Savannah River Site, DOE began to blend excess fuel-grade plutonium from N reactor with super-grade from the Savannah River Site to produce weapons-grade plutonium. (Refs. 31 & 128)

1982: The Savannah River Site heavy water plant stopped deuterium production. (Ref. 128)

1982: Nuclear Waste Policy Act. (Ref. 128)

1982: The name of the Lawrence Livermore Laboratory (LLL) was changed to the Lawrence Livermore National Laboratory. (Ref. 128)

1982: Plutonium laser isotope separation was demonstrated at Lawrence Livermore National Laboratory. (Ref. 128)

February 1983: Strategic Defense Initiative Announced. (Ref. 128)

April 1983: Leaf v. Hodel Decision Subjects DOE to RCRA. (Ref. 128)

1983: PUREX Plant at Hanford resumes operation. After restart, a new line in the PUREX N-Cell was used to convert plutonium nitrate solutions to more stable plutonium oxide. The plutonium oxide was transferred to the plutonium finishing plant (PFP) in the Hanford 200 West are for conversion to metal. (Refs. 19, 28 & 128)

1983: The processing facility at Tomsk-7 replaces the acetate precipitation process with the Purex process. (Refs. 24, 25, 26 & 27)

1984: At Hanford's Plutonium Finishing Plant the remote mechanical line for conversion of plutonium nitrate and plutonium oxide to metal in restarted and operates intermittently until 1990. (Refs. 28 & 128)

1985: L Reactor at Savannah River was restarted. (Ref. 128)

1985: AKSRS, ²³⁸Pu recovery operations shifted to the new HB-Line. (Ref. 128)

1985: Due to an accident at PFP, plutonium oxide from Hanford was sent to LANL TA-55 for conversion to metal for several months. (Refs. 28 & 128)

1986: C-Reactors stopped operating, than went on cold standby in 1987 after engineers determined that cracks in the reactor vessel, discovered in 1984, could not be fixed. (Ref. 128)

April 26, 1986: Chernobyl Nuclear Disaster. At the time of the accident Chernobyl Unit-4 was operating at an average burn-up of 10.3 gigawatt days per metric ton of uranium. (Refs. 128, 130 & 132)

-64- 4/15/99

1987: Uranium enrichment plant at K-25 was shutdown completely. (Ref. 128)

1987: Fuel and target fabrication at Hanford's 300 area ceased permanently. (Ref. 128)

1987: N Reactor at Hanford is shut down permanently. The nine reactors at Hanford had produced a total of 67.4 metric tons of plutonium including 54.5 metric tons of weapons-grade plutonium. The reactors had also produced tritium, polonium-210, uranium-233, thulium-170, iridium-192, and other special use isotopes. (Refs. 28 & 128)

December 1987: Intermediate-Range Nuclear Force Treaty (INF). (Ref. 128)

February 1988: Secretary of Energy John Herrington tells a congressional subcommittee the United States is "awash in plutonium." (Refs. 19 & 128)

June 1988: L Reactor at the Savannah River Site is shut down for safety upgrades. (Refs. 19 & 128)

August 1988: P Reactor at the Savannah River Site is shut down for safety upgrades. (Refs. 19 & 128)

December 1988: PUREX Plant at Hanford shut down for terminal cleanout. (Refs. 19, 28 & 128)

May 1989: Hanford Tri-Party Agreement. (Refs. 28 & 128))

June 1989: Rocky Flats Plant raided by FBI. (Ref. 128)

1989: A National Academy of Sciences panel, using classified data, concluded that additional plutonium production is unnecessary. (Ref. 19)

1989: The US DOE nominated the "B" reactor at Hanford to the National Register of Historic Places (on an emergency basis, because it is less than 50 years old). (Ref. 28)

July 1989: Processing of recycled uranium at the Feed Materials Production Center at Fernald is ended. (Ref. 128)

December 1989: The Rocky Flats Plant was shut down to bring it into compliance with environmental regulations. However, the plant's defense mission was cancelled in 1992 due to a change in the needs of the nuclear weapons stockpile. Workers at Rocky Flats loaded the last plutonium "trigger" for a nuclear warhead into a tractor trailer bound to the Pantex Plant. The nuclear weapon built with this trigger was the last one made in the United States for the foreseeable future. (Refs. 19 & 128)

December 1989: The FB-Line at the Savannah River Site shut down for maintenance. (Ref. 128)

-65- 4/15/99

1990: Hanford's PUREX closed permanently after a short clean-out run. It had operated intermittently in the late 1980's. (Refs. 19, 28 & 128)

1990: At Hanford's Plutonium Finishing Plant the remote mechanical line for conversion of plutonium nitrate and plutonium oxide to metal in shut down. (Refs. 28 & 128)

1990: The Soviet Union collapses bringing the nuclear arms race of the "Cold War" to a sudden end. (Refs. 128 & 131)

1990: All available, blendable N-Reactor-produced fuel-grade plutonium blended at Savannah River to weapons-grade plutonium. (Ref. 128)

July 31, 1991: START I signed in Moscow by the United States and the Soviet Union. START I was the first arms control treaty to enter into force that mandated reductions of deployed strategic weapons as opposed to limitations of future deployments. (Ref. 131)

1991: The Soviet Union dissolves. (Refs. 19 & 128)

September 1991: The F-canyon at Savannah River is shut down as the result of safety concerns. The H-Canyon also shut down in response to the Secretary of Energy's determination to discontinue spent fuel reprocessing. (Ref. 128)

January 28, 1992: President George Bush announces in his State of the Union Address the decision to cancel all new submarine launched ballistic missile warhead production. He also announced that the remaining planned production of the W88 warhead was canceled. President Bush also announced that with this cancellation, plutonium components (pit) manufacturing operations at the RFP are terminated at this time. The State of the Union Address also offered a bilateral plan for further reduction in nuclear weapons, provided there was a reciprocal response by the Commonwealth of Independent States (CIS). (Ref. 131)

1992: The United States stopped nuclear explosion testing. (Ref. 131)

1992: The ICPP at INNEL shuts down. During its operation, it had recovered a total of 31.5 tonnes of high-enriched uranium from naval (5.1 tonne) research and test reactor fuel. (Ref. 128)

1992: Congress passed the Energy Policy Act and, under its provisions, DOE leased uranium enrichment operations at the Portsmouth and Paducah Plants to the newly created United States Enrichment Corporation (USEC). (Ref. 128)

July 1992: K-Reactor at Savannah River was placed in standby. While the main products of the Savannah River reactors were weapons grade plutonium (a total of 36.1 metric tons) and tritium, a variety of isotope products including uranium-233, americium-243, curium-244, polonium-210, cobalt-60, plutonium-238, plutonium-242, and californium-252 were also produced. (Ref. 128)

September 1992: Last US Nuclear Test — code named "Divider." (Refs. 19 & 128)

-66- 4/15/99

November 1992: The Russian parliament approved START I, with the condition that Russia would not exchange instruments of ratification until Belarus, Kazakstan, and Ukraine acceded to the NPT as non-nuclear-weapons states. (Ref. 131)

1992: Two plutonium production reactors (AD and ADE-1) are closed down at Krasnoyarsk-26. According to various estimates krasnoyarsk-26 produced more than 45 metric tons of weaponsgrade plutonium as plutonium dioxide. (Refs. 3, 25, 26 & 27)

January 1993: START II Treaty signed by Presidents Bush and Yeltsin. (Refs. 128 & 131)

April 6, 1993: Chemical explosion in the radiochemical processing plant of the Siberian Chemical Combine in Tomsk-7. The explosion caused substantial damage to the crane bay above the process area, blew out several hundred feet of non-reinforced masonry wall and a section of the roof, wrecked internal components, and started small roof fires. The accident contaminated an area of about 123 square kilometers inside and outside the plant with approximately 40 curies of radioactive material. (Ref. 24)

May 5, 1993: Russian Minister of Atomic energy, Mr. Viktor Mikhailov extended an invitation to US DOE Secretary O'Leary to have a contingent of DOE experts visit Russia, and in particular the Tomsk-7 site, for an informational exchange meeting. (Ref. 24)

June 23 – 26 1993: Contingent of DOE scientist tours the radiochemical processing area at Tomsk-7 and engage in discussion concerning the explosion. (Ref. 24)

1993: The third Savannah River Site tritium facility began operations. (Ref. 128)

September 27, 1993: President Clinton announced the "Nonproliferation and Export Control Policy which states: "The United States does not encourage the civil use of plutonium and accordingly, does not itself engage in plutonium reprocessing for either nuclear power or nuclear explosive purposes. The United States, however, will maintain its existing commitments regarding the use of plutonium in civil nuclear programs in Western Europe and Japan." (Refs. 31 & 80)

January 1994: US President Clinton and Russia's President Yeltsin issued a Joint Statement Between the United States and Russia on Nonproliferation of Weapons of Mass destruction and Means of their Delivery. In this joint statement the Presidents tasked their experts to jointly "study options for the long-term disposition of fissile materials, particularly of plutonium, taking into account the issues of nonproliferation, environmental protection, safety, and technical and economic factors. (Ref. 80)

May 1994: Russia agreed to cease operating two plutonium production reactors at Tomsk-7 and one at Krasnoyarsk-26 by the year 2000, with the expectation that alternative sources of energy would be available to these cities by that date to substitute for the district heating the reactors provide. The agreement was formalized at the June 1994 meeting of the Gore-Chernomyrdin Commission (GCC) Russia has refused to bring the agreement into force. (Ref. 3)

December 5, 1994: START I entered into force. Now being implemented, it includes verified destruction of strategic launchers (silos, bombers, submarines) and data exchanges regarding current deployments of strategic weapons. Total Russian and U. S. strategic arsenals are to be reduced to 6,000 accountable warheads by 2002. (Ref. 3)

March 1, 1995: President Clinton announces that 200 metric tones of US fissile materials had been declared surplus to the US defense needs. (Refs. 31 & 80)

June 22, 1995: Russian President Yeltsen submitted the START II treaty to the Duma; prospects for treaty ratification are uncertain due to domestic and international developments. (Ref. 3)

1995: Mound ends its production activities. (Ref. 128)

1995: Pinellas ends its production activities. (Ref. 128)

January 26, 1996: START II approved for ratification by the US Senate. START II will cap the number of deployed strategic warheads at 3,500.

February 6, 1996: The US DOE declassified and disclosed the location and form of all US excess fissile material, amounting to 20% of the total US stocks of plutonium and highly enriched uranium. (Ref. 31)

1996: The defense Waste Processing Facility (DWPF) at the Savannah River Site begins operation. It is anticipated that approximately three decades will be necessary to convert approximately 34 million gallons of high-level waste into thousands of glass logs. (Ref. 128)

1996: F-Canyon at Savannah River Site restarted to stabilize nuclear materials. (Ref. 128)

1996: Non-Proliferation Treaty Renewed. (Ref. 128)

1996: UN Comprehensive Test Ban Approved. (Ref. 128)

January 14, 1997: The United States Department of Energy announced a hybrid disposition strategy which calls for immobilizing surplus plutonium with high level waste and using surplus plutonium as mixed oxide (MOX) fuel in existing commercial reactors to render the materials non-weapons-usable. (Ref. 125)

-68- 4/15/99